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Joint Research Centre

Experimental Measurements of Prompt Fission Neutrons and γ -rays

F.-J. Hamsch

*FIESTA2017 School and Workshop,
Santa Fe, NM, Sept. 18 – 22, 2017*



Table of content

- **General aspects of neutron and γ -emission**
- **Prompt neutron measurements**
- **Prompt γ -ray measurements**

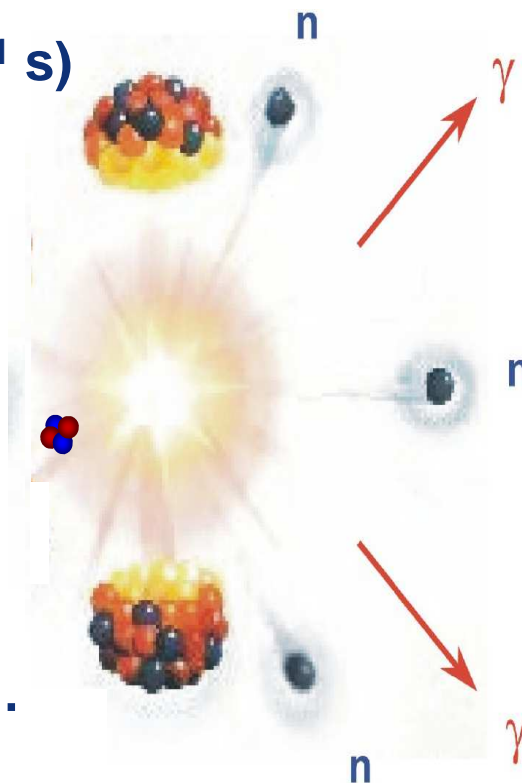
The fission process

prompt neutrons (10^{-18} s)

prompt γ -rays (10^{-16} s)

fission fragments (10^{-21} s)

ternary α , t, d, ^{10}Be ...



kinetic energy
prompt γ -rays

heat

prompt neutrons
(delayed neutrons)

chain reaction

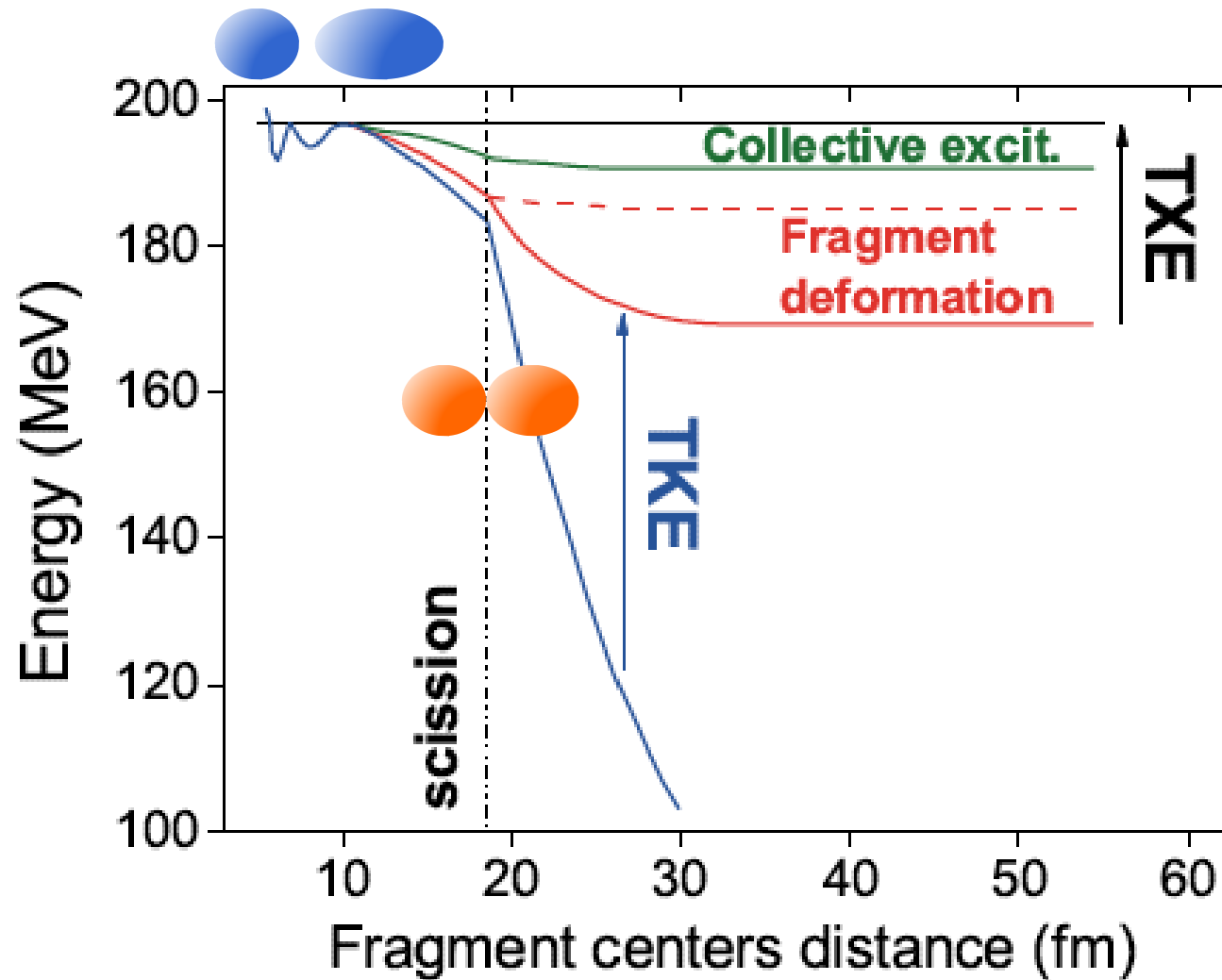
ternary α , t, d

gas production
in the fuel (waste)

fission fragments

decay heat,
toxicity (waste)

The fission process



Fission fragment de-excitation

- **Excitation energy of the fragments is dissipated through particle emission, here essentially neutrons and γ -rays**
- **On average 2 – 4 neutrons are released**
- **The exact value depends on the isotope and the excitation energy of the compound nucleus e.g. for $^{235}\text{U} \sim 2.4$)**
- **The average energy of a neutron in the LS is around 2 MeV**
- **On average 6 – 10 γ -rays are emitted too, with a mean total energy release of about 7 – 9 MeV**

Fission fragment de-excitation

➤ Observable quantities:

- **Spectral characteristics (neutrons and γ -rays)**
 - **Average multiplicity** (/fission)
 - **Average total energy** (/fission)
 - **Average photon energy** (/fission)
- **Correlations with fission fragment characteristics**
 - $\nu(A^*, \text{TKE}), \langle E_{\text{tot}} \rangle(A^*, \text{TKE}),$
- **Correlation of prompt γ -ray data with PFN**

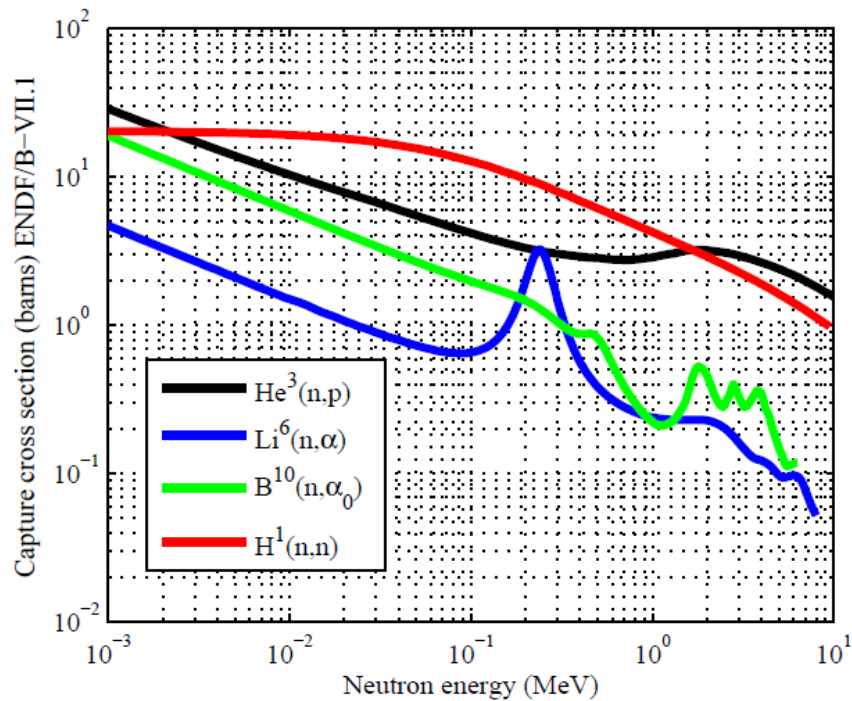
How to measure neutrons and γ -rays

➤ Prompt neutron measurements

How to measure neutrons

- **Suitable detectors**
- **Any material with a high capture cross section for neutrons (H, He, Li, B)**
 - **He-3 counters**
 - **Boron (BF_3) counters**
 - **Lithium-glass detectors**
 - **Liquid scintillator detectors (mainly containing H)**

Choice of material (large interaction cross section)



PhD thesis, A. Riego, UPC Barcelona (2016)

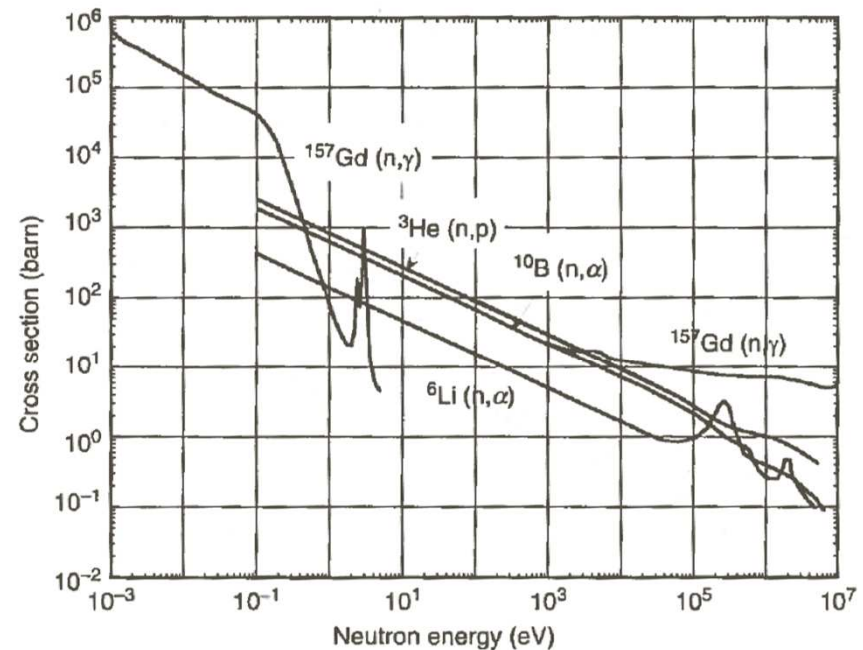


Figure 3.30 Neutron cross sections for several isotopes. Data from Garber and Kinsey (1976).

F. Sauli, Gaseous Radiation Detectors, Cambridge U. Press, 2014

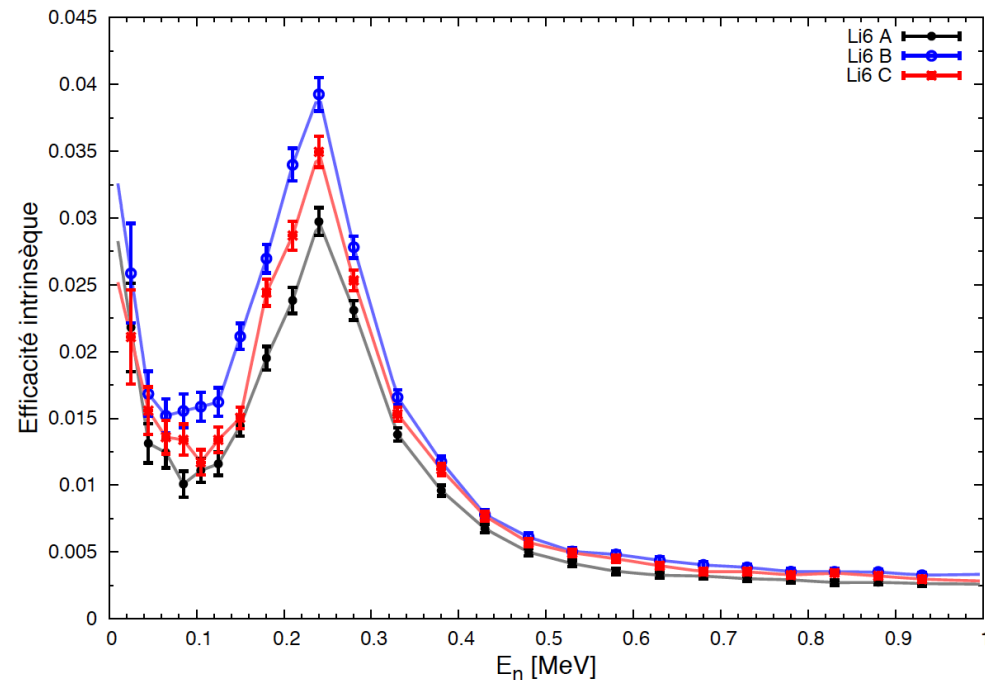
How to measure neutrons

➤ Lithium-glass detectors:

- Enriched in ^6Li : $^6\text{Li}(n,t)\alpha$
- Exothermic reaction: $Q = 4.8 \text{ MeV}$

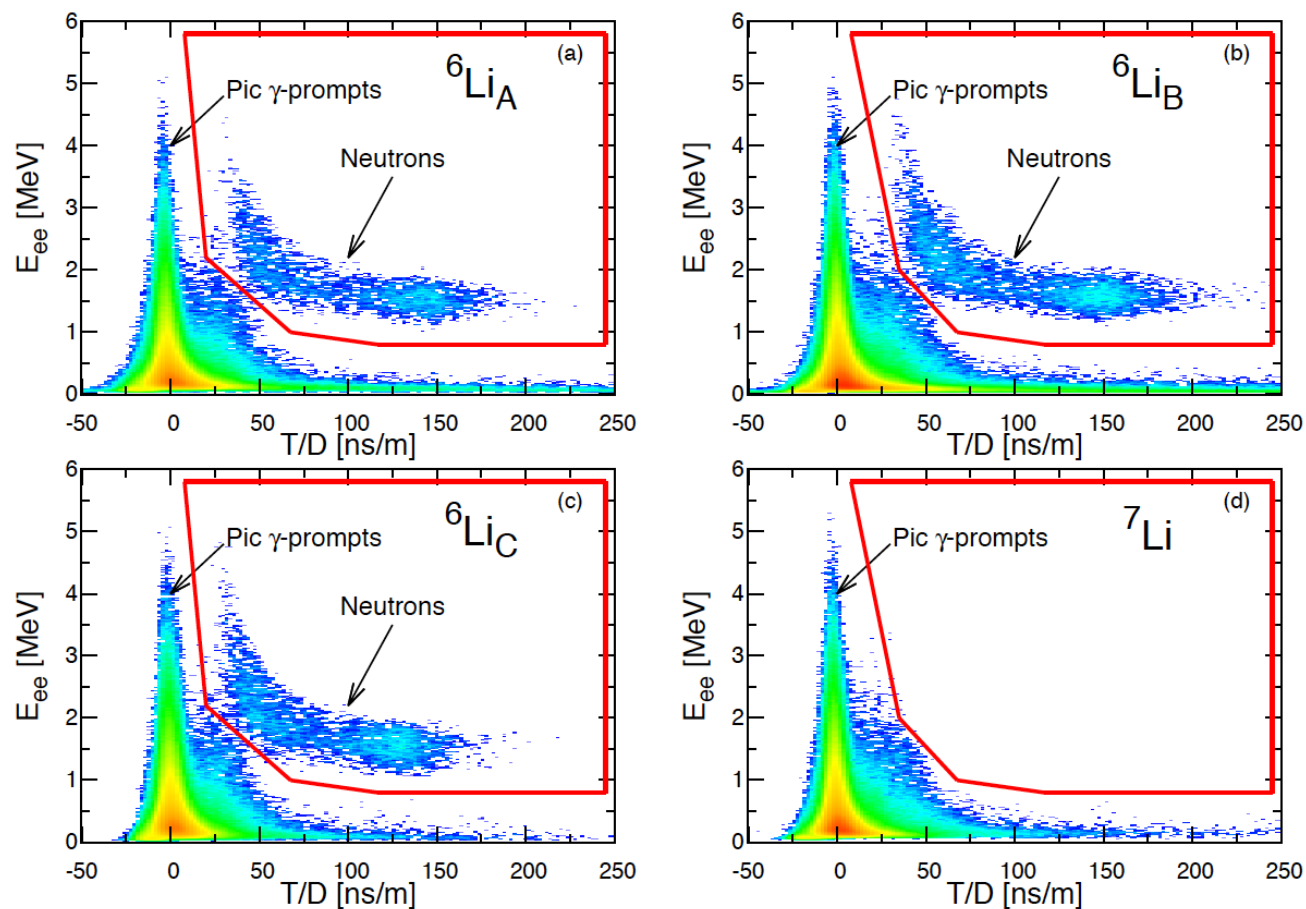
➤ CONS:

- Relatively low detection efficiency
- Bad timing resolution prevents from using longer crystals



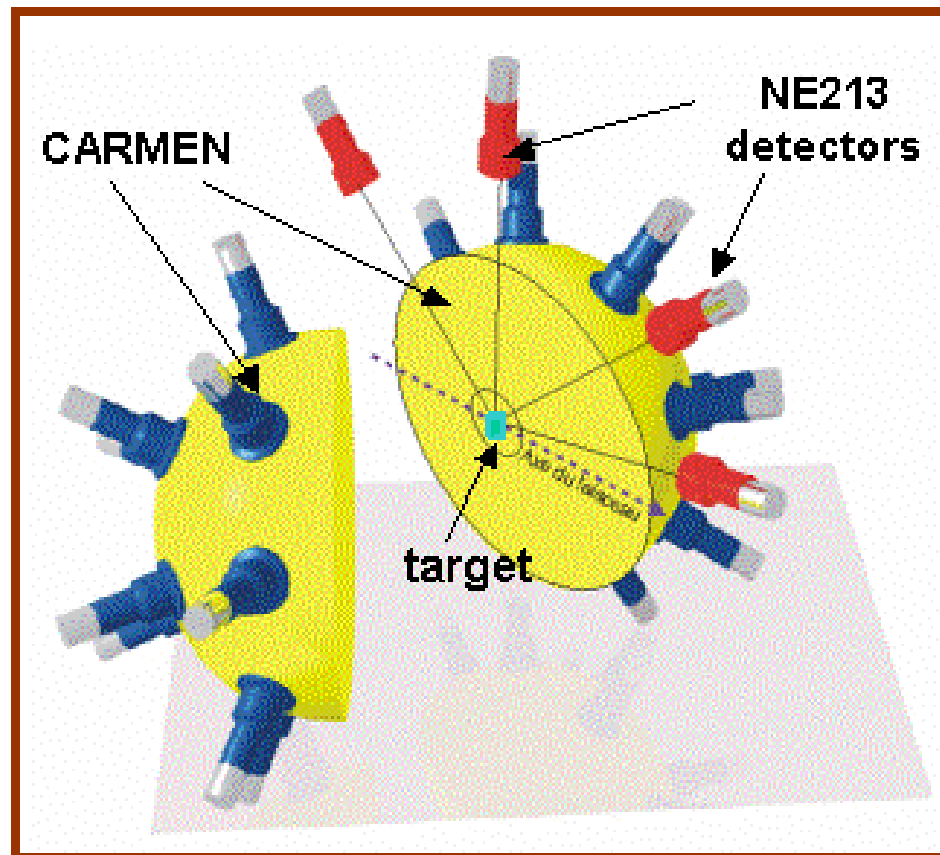
How to measure neutrons

➤ Lithium-glass detectors:



How to measure neutrons

- **Detectors based on liquid scintillator(s):**
 - **Gd loaded scintillation tanks**
 - **NE213 equivalent scintillators**



Liquid scintillation tank

➤ Pros:

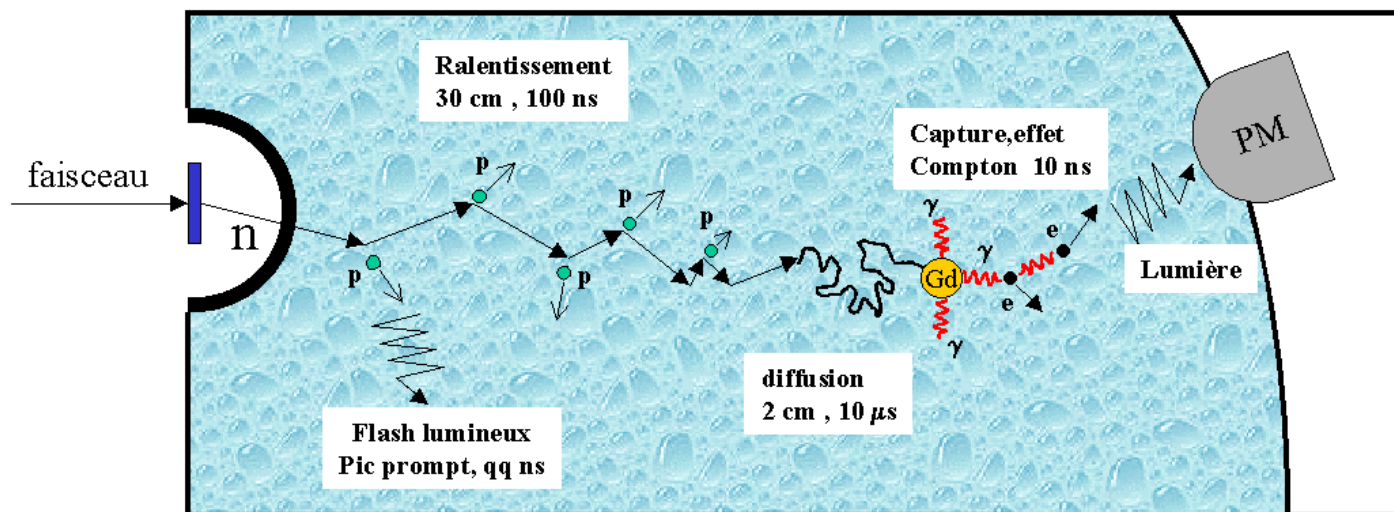
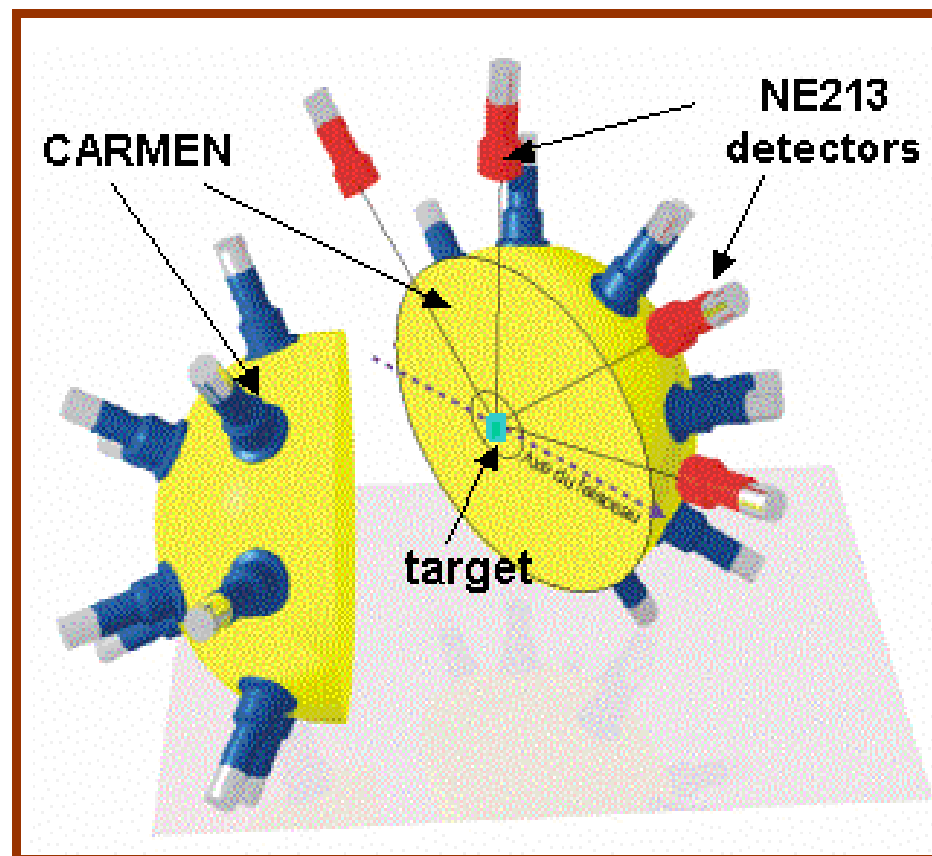
- High efficiency (4π geometry)
- Large thermal neutron capture cross section of Gd isotopes
- Release of a high energy gamma cascade

➤ Cons:

- High toxicity (xylene- or dioxane-based) scintillator
- Low flash point -> highly flammable

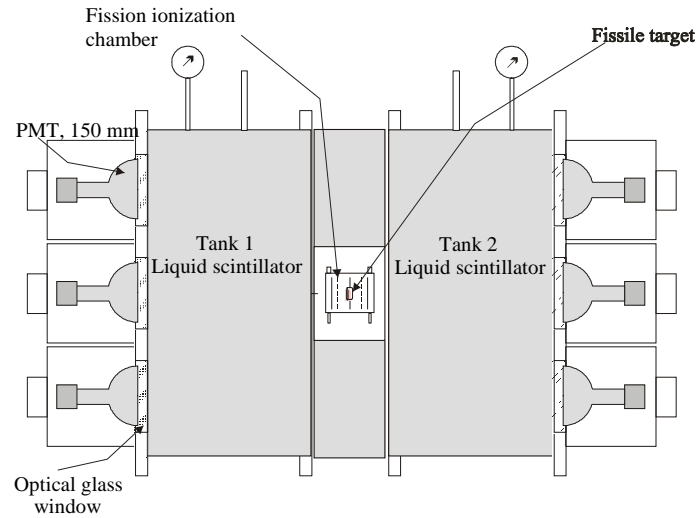
Liquid scintillation tank (CEA-Arpajon)

- 85 % eff for fission neutrons
- Two hemispheres (r=60cm)
- 950l (C_9H_{12}) with 0.5% Gd
- 24 phototubes
- Can be combined with other Detectors (e.g. NE213)

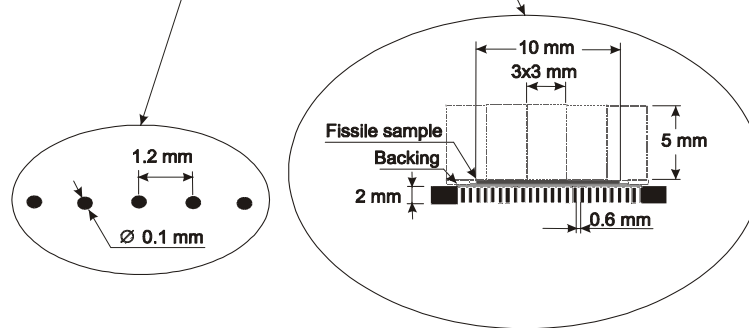
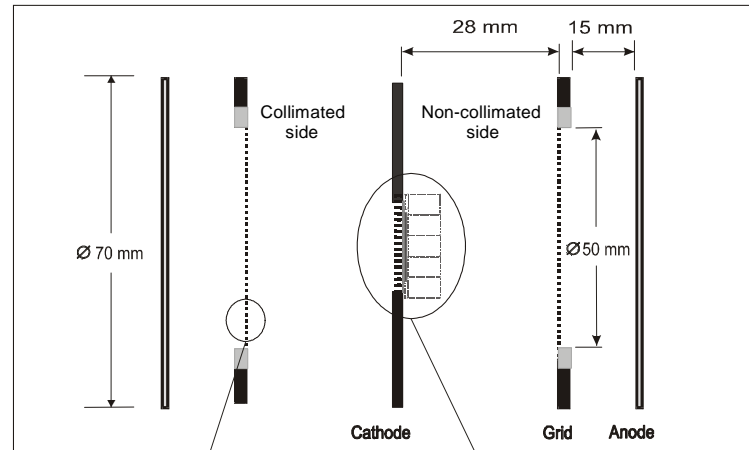
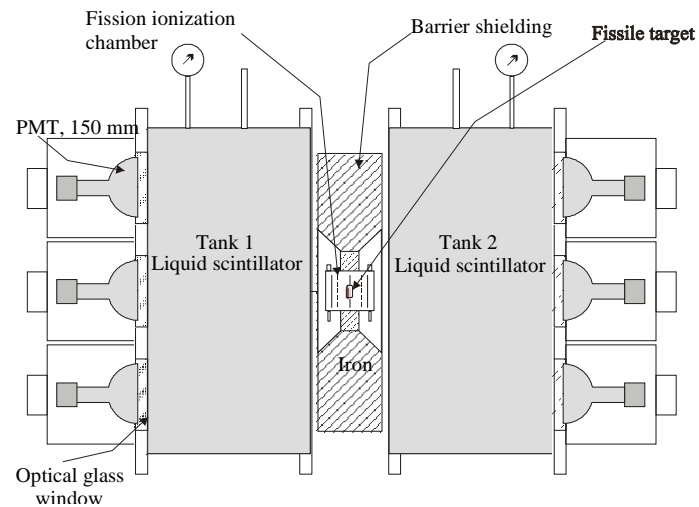


Liquid scintillation tank (used at PNPI, Gatchina, Russia)

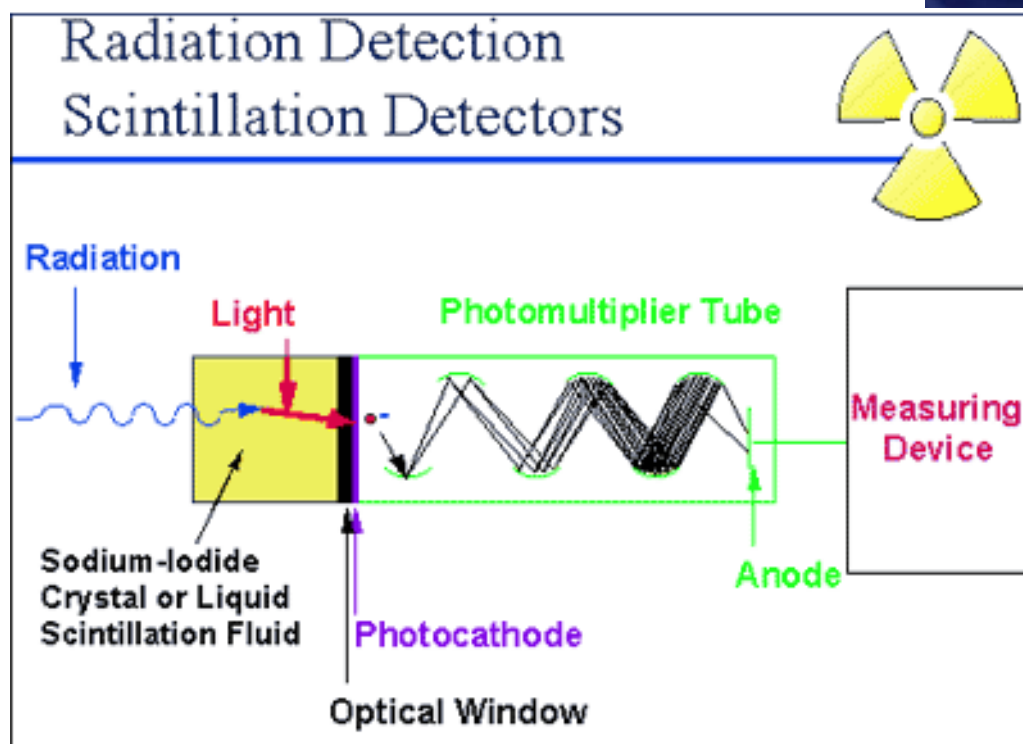
4 π -geometry



2 x 2 π - geometry



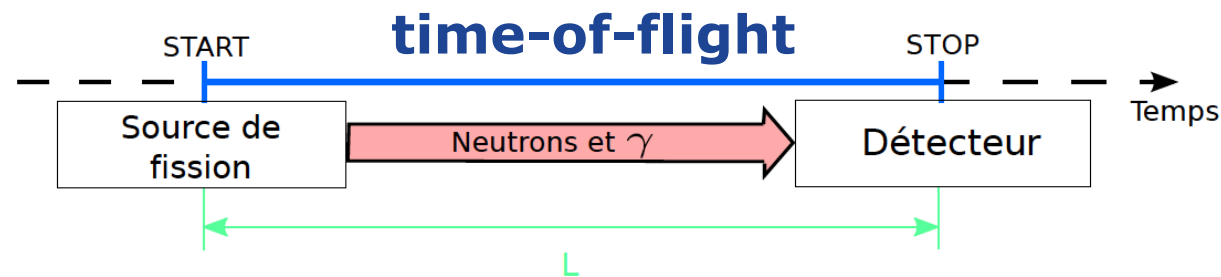
Scintillation detectors and photomultipliers



Source: WWW

How to measure neutrons

➤ Prompt neutron measurements



$$E_n = (\gamma - 1)m_n c^2 = \left(\frac{1}{\sqrt{1 - \frac{L^2}{\Delta t^2 c^2}}} - 1 \right) m_n c^2$$

➤ $m_n = 939.56533 \text{ MeV}/c^2$

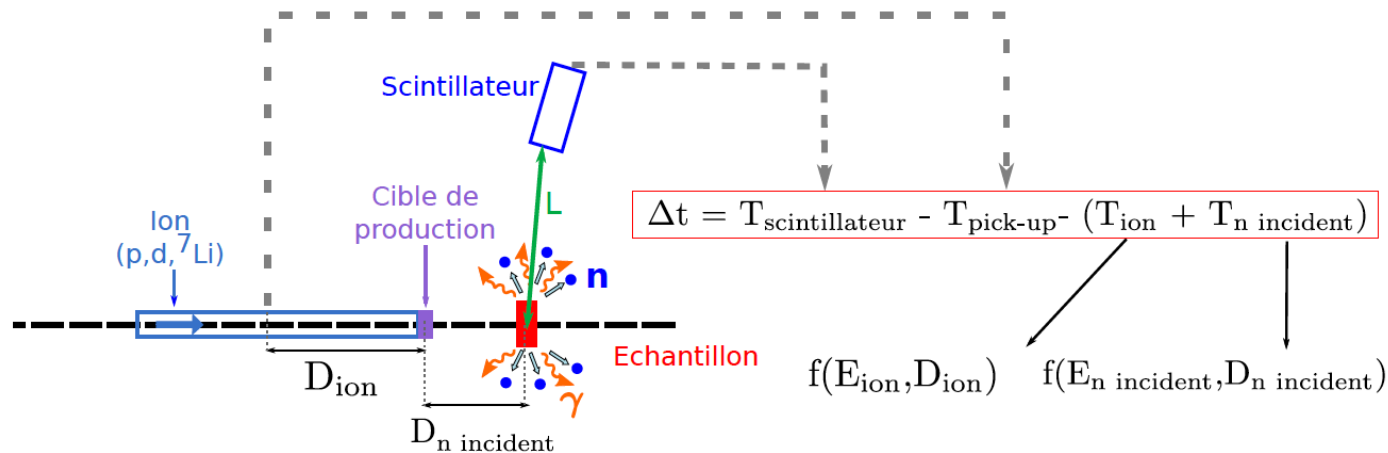
➤ $c = 0.299792458 \text{ m/ns}$

➤ Δt : time of flight (TOF)

$$\left(\frac{\sigma_E}{E} \right)^2 = 2 \left[\left(\frac{\sigma_L}{L} \right)^2 + \left(\frac{\sigma_{\Delta t}}{\Delta t} \right)^2 \right]$$

How to measure neutrons

➤ Measurement with passive sample



- Use of massive targets (several g)
- Pulsed neutron beam (usually low beam currents; 1 – 2 μA)
- In general leads to a sufficiently high event rate
- Resolution depends on beam pulse
- Minimum neutron energy depend on incident neutron energy
- Multiple scattering in the sample

How to measure neutrons

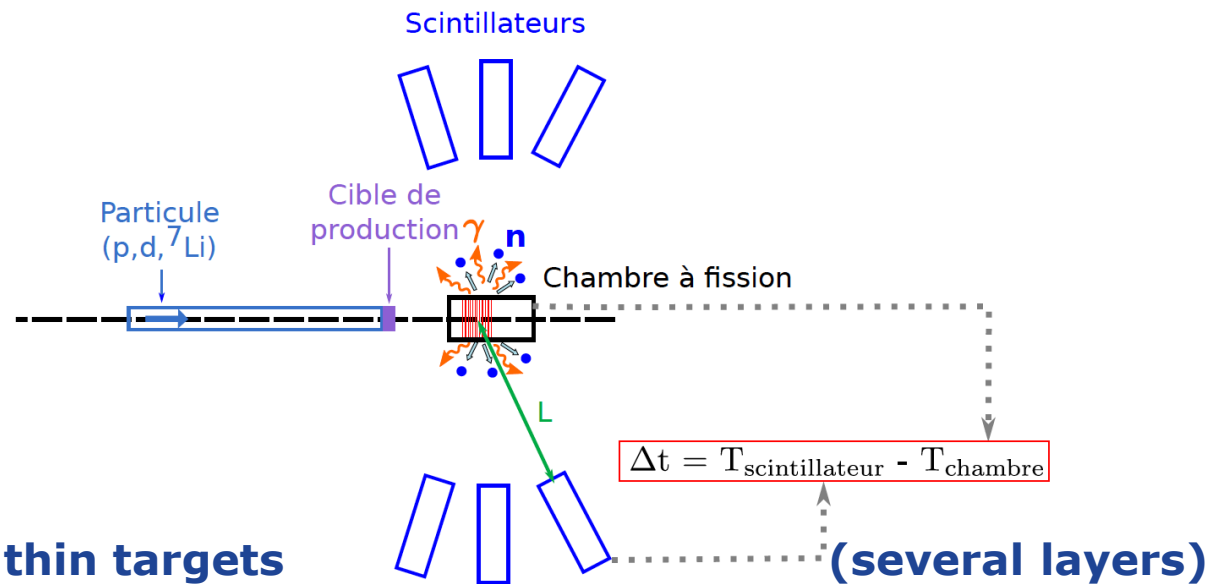
➤ The measurement environment (direct reaction):



- Limited number of detectors
- Contributions from neutron scattering
- Simulation by means of MCNP or Geant4

How to measure neutrons

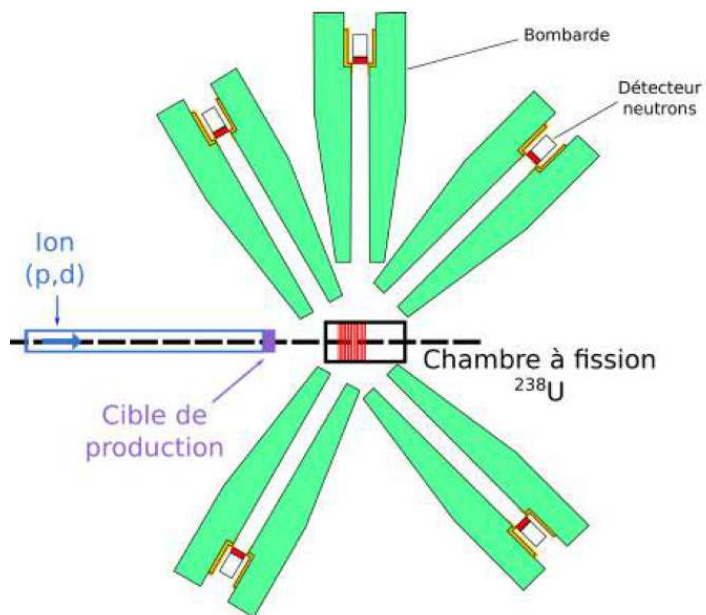
➤ Measurement with an active sample



- Use of thin targets
- Continuous neutron beam (high beam currents; $> 20 \mu\text{A}$)
- Allows to measure neutrons below the beam energy
- Allows measuring at different energies with changing particle beam
- Multiple scattering in the detector to be taken care of

How to measure neutrons

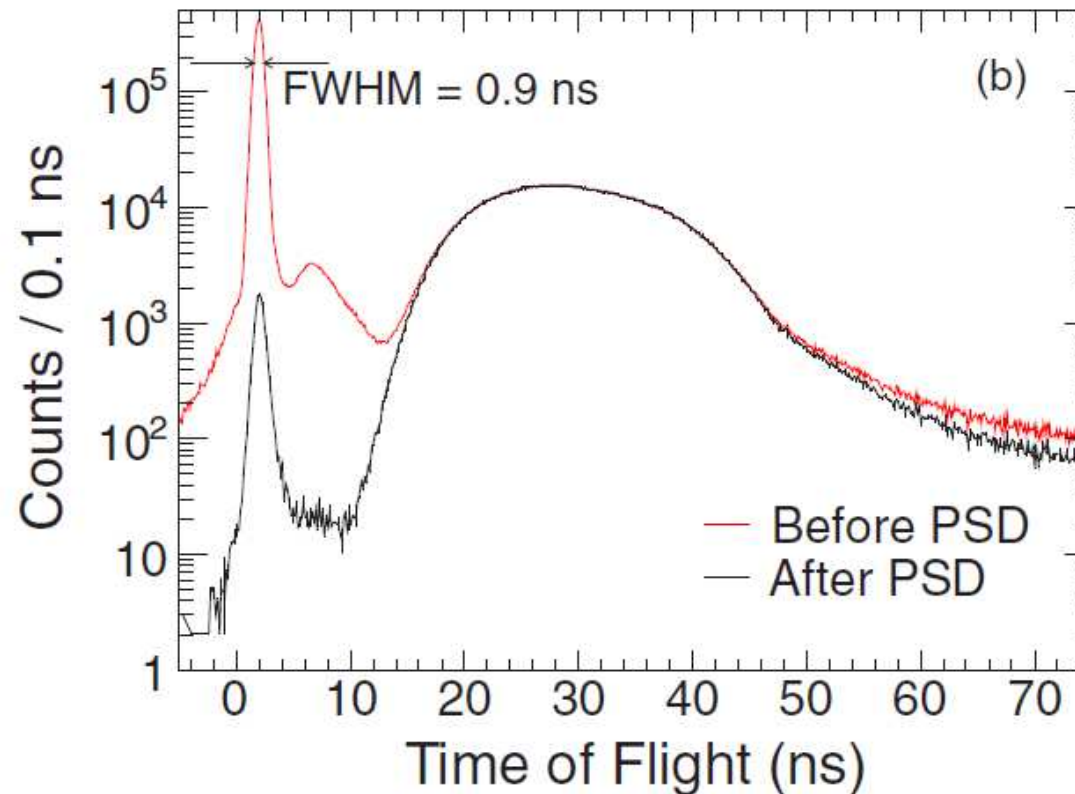
➤ The measurement environment (direct reaction):



- Limited number of detectors
- Contributions from neutron scattering...

How to measure neutrons

- Detectors based on liquid scintillator(s):
 - Very fast detectors: $\sigma_t < 1$ ns
 - Neutron – γ separation by means of TOF



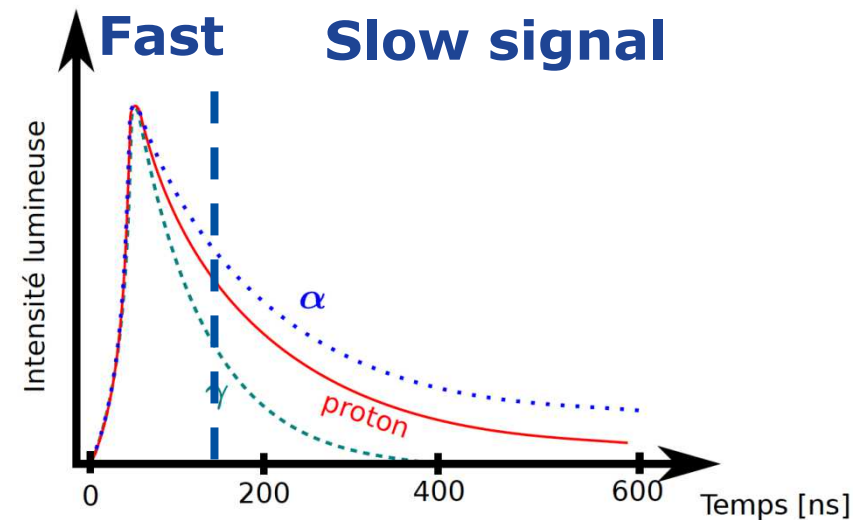
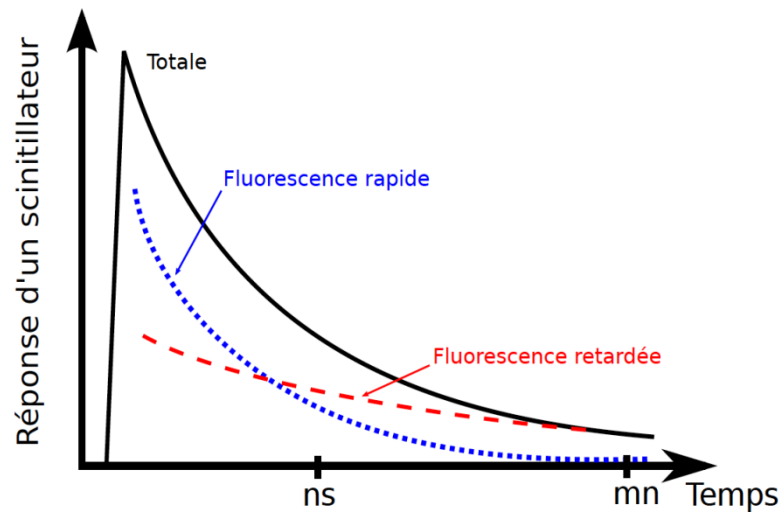
How to measure neutrons

➤ Detectors based on NE213 liquid scintillator(s):

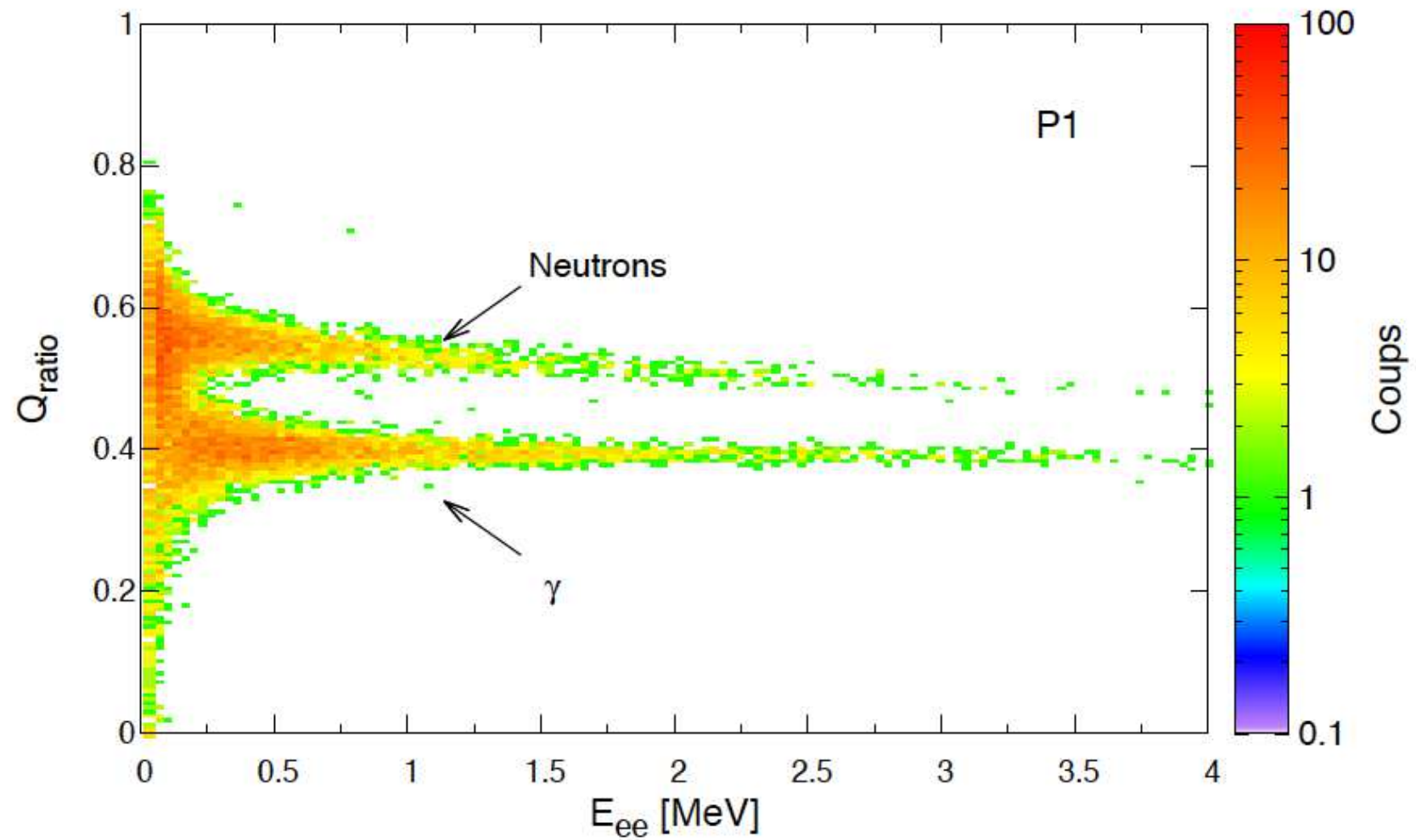
➤ Allow pulse shape discrimination

➤ Electrons and recoil protons excite different fluorescent levels

➤ Detector signal shows different fall times



How to measure neutrons



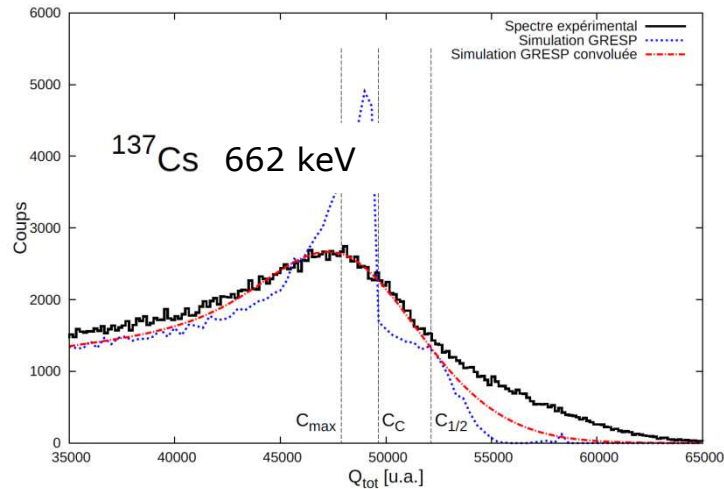
PhD thesis, A. Sardet, Université Paris Sud (2015)



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Detector calibration

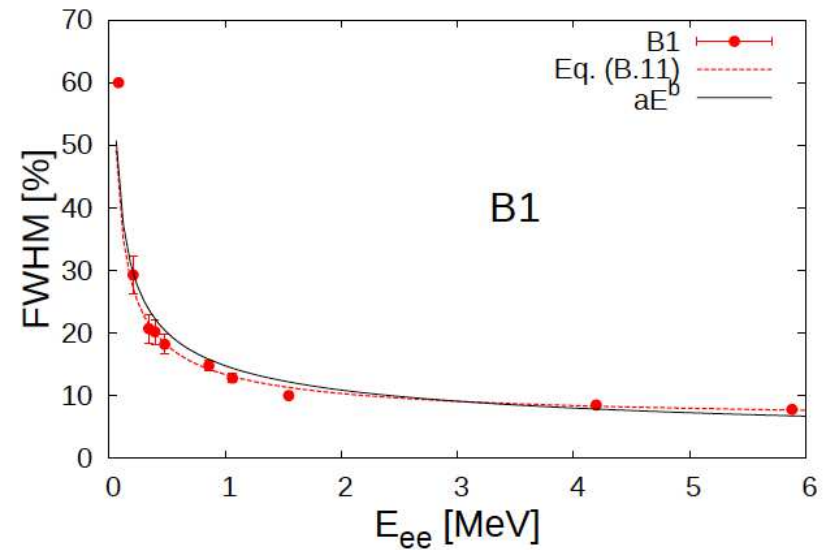
➤ Response of LS detectors to neutrons and γ -rays:



$$\frac{\Delta E}{E} = 1.5 \frac{C_{1/2} - C_{\text{max}}}{C_{1/2}}$$

Simulation with GRESP

$$\frac{\Delta E}{E} = \sqrt{\alpha^2 + \frac{\beta^2}{E} + \left(\frac{\gamma}{E}\right)^2}$$



A. Sardet, et al., NIM A792 (2015) 74

PhD thesis, A. Sardet, Université Paris Sud (2015)

Detector calibration

➤ Selection of γ -rays sources for calibration

Source	E_{γ} [keV]	E_{ee} [keV]	Type	Résolution B1 [%]
^{133}Ba	81	81	FE	$59,98 \pm 0,2$
^{133}Ba	356	207	CE	$29,27 \pm 3,2$
^{22}Na	511	340,7	CE	$20,7 \pm 2,3$
^{207}Bi	569	393,3	CE	$20,2 \pm 2,0$
^{137}Cs	662	477,65	CE	$18,2 \pm 1,6$
^{207}Bi	1063	857,7	CE	$14,8 \pm 0,9$
^{22}Na	1 275	1 061,7	CE	$12,8 \pm 0,7$
^{207}Bi	1770	1 546,9	CE	$10,0 \pm 0,6$
AmBe	4 430	4 196	CE	$8,5 \pm 0,4$
Pu^{13}C	6 130	5 883	CE	$7,8 \pm 0,3$

➤ Use of mono-energetic neutron beams for calibration

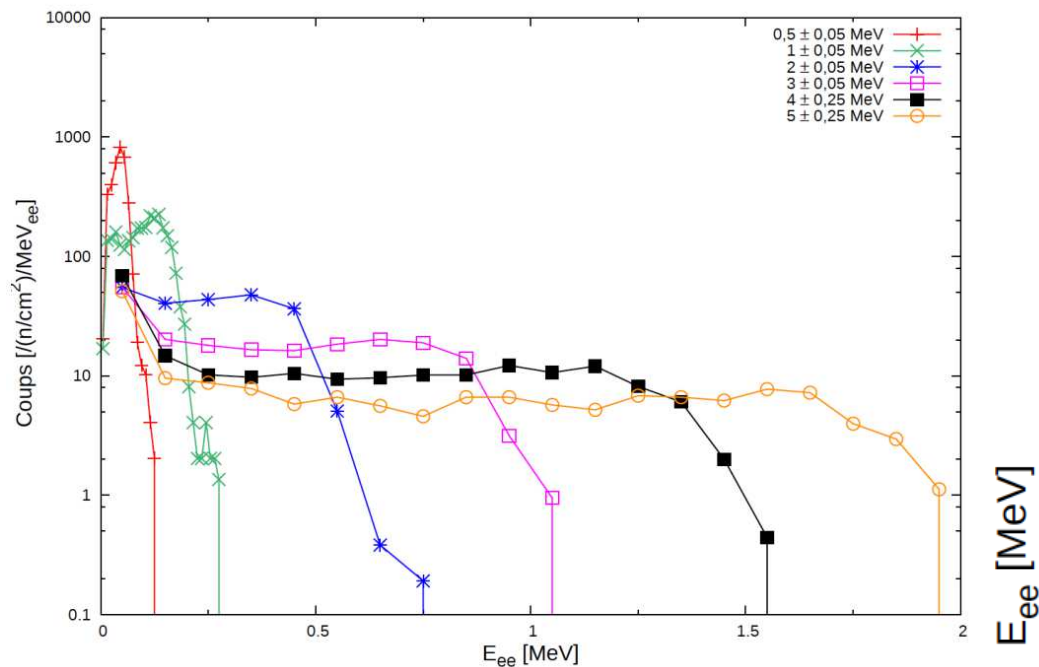
CE: Compton Edge

FE: Photo peak

PhD thesis, A. Sardet, Université Paris Sud (2015)

Detector calibration

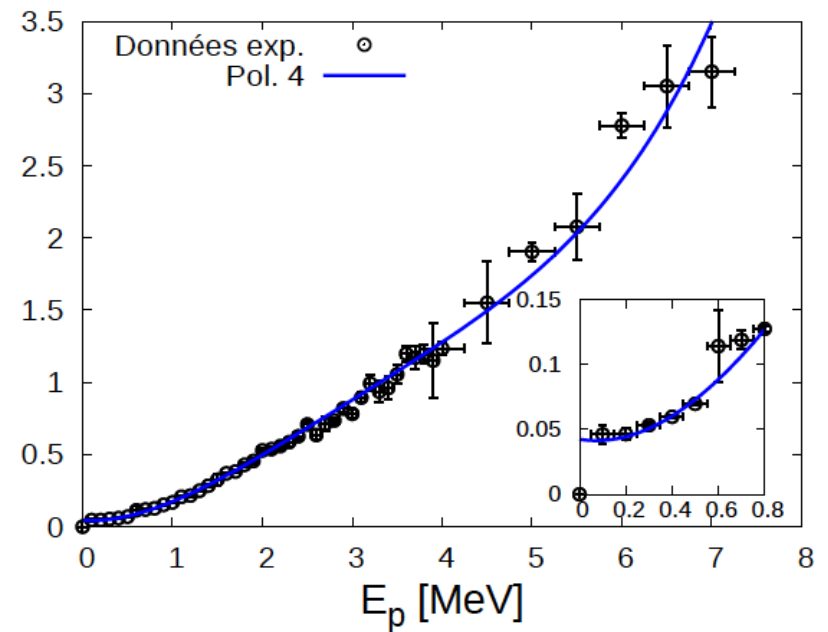
➤ Response of LS detectors to neutrons and γ -rays:



➤ Response for mono-energetic neutrons

➤ Selection from the TOF information

➤ Calibrated neutron beam

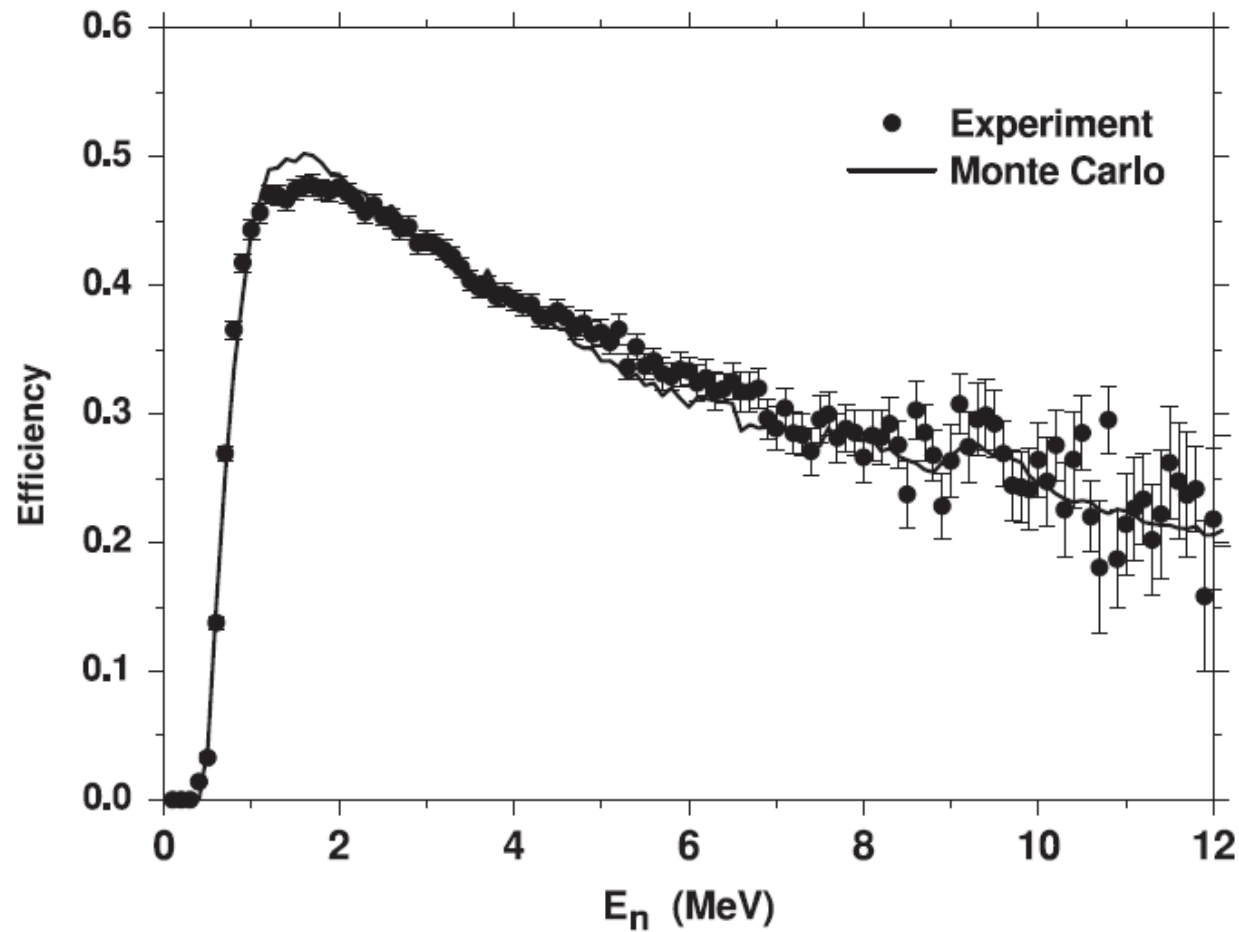


A. Sardet, et al., NIM A792 (2015) 74

PhD thesis, A. Sardet, Université Paris Sud (2015)

Efficiency curve

➤ Response of LS detectors to neutrons and γ -rays:



How to measure neutrons

➤ And finally: a prompt fission neutron spectrum

$$dN/dE \sim Y \Omega \varepsilon \nu S(E) C_n$$

Y : Fission yield

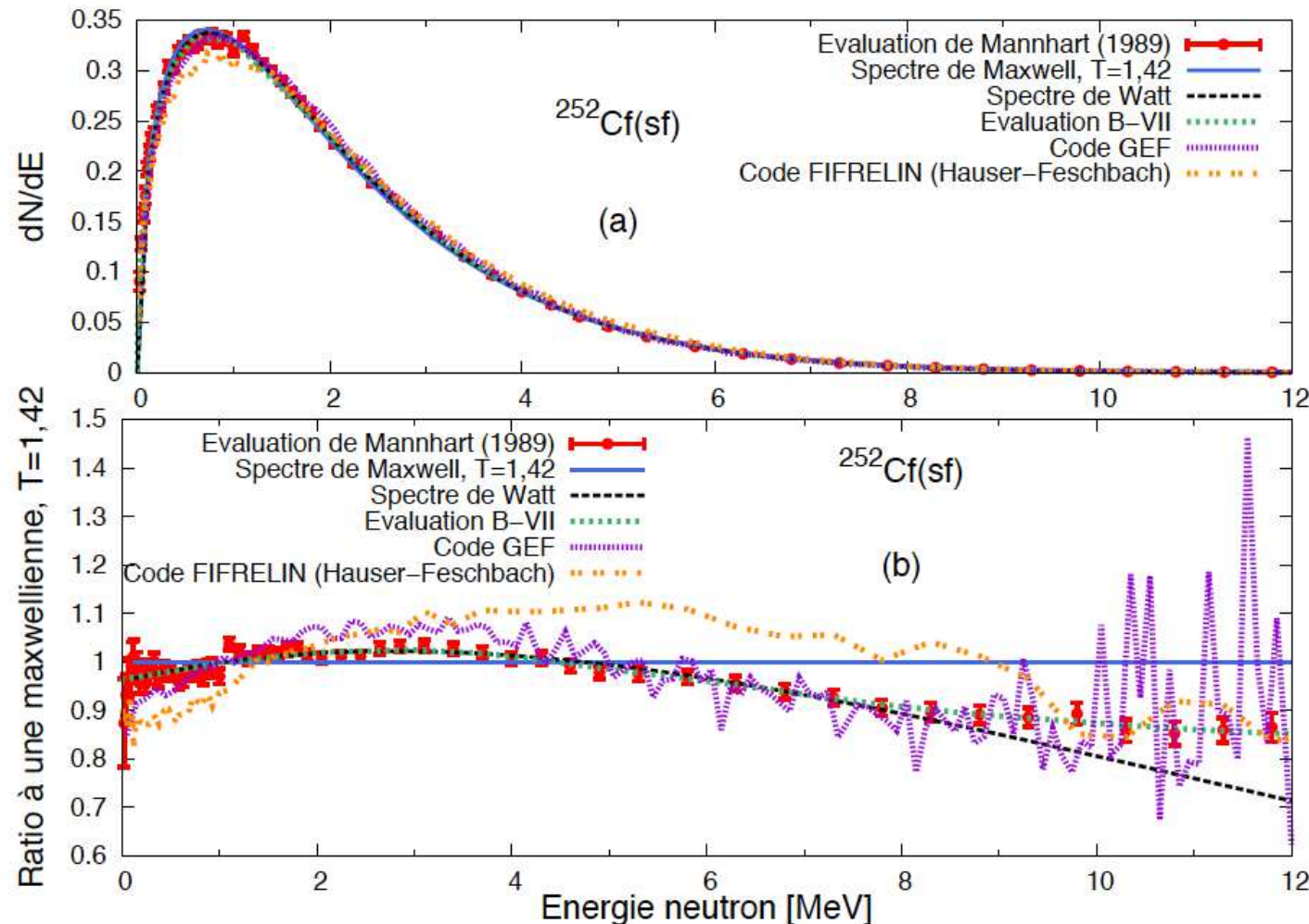
Ω : Solid angle

ε : Efficiency

ν : n-multiplicity

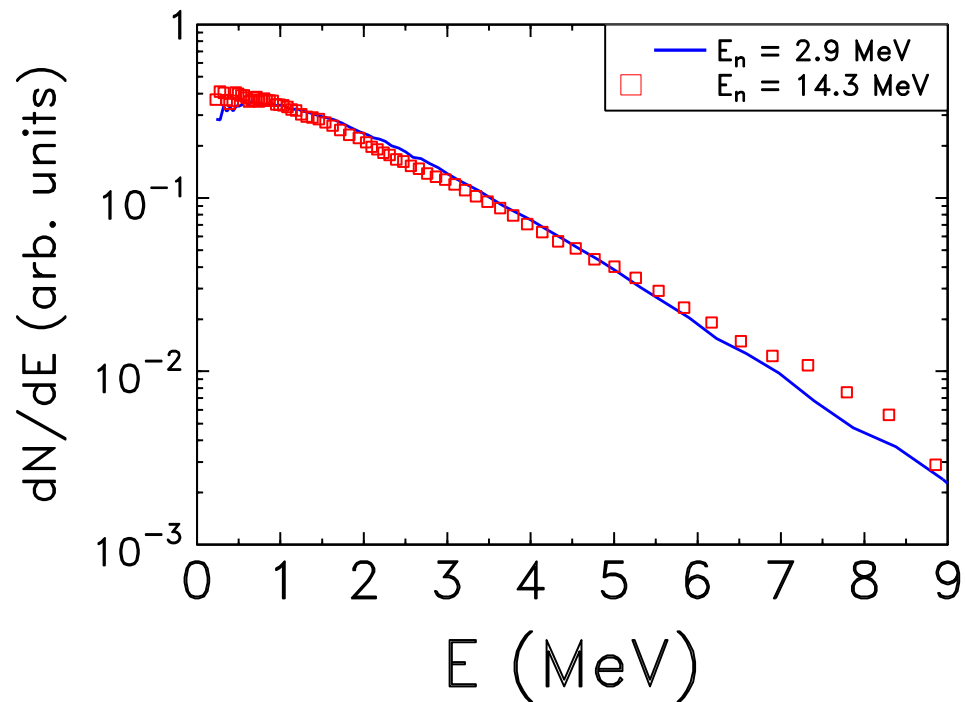
S(E) : Spectrum
from ToF

C_n : correction factors

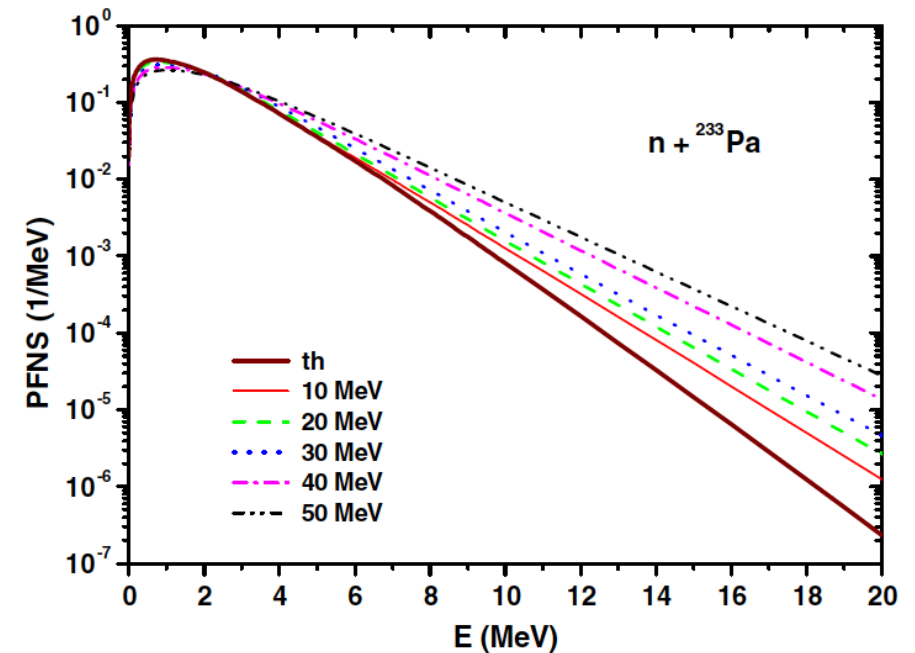


How to measure neutrons

➤ Energy dependence of PFN emission



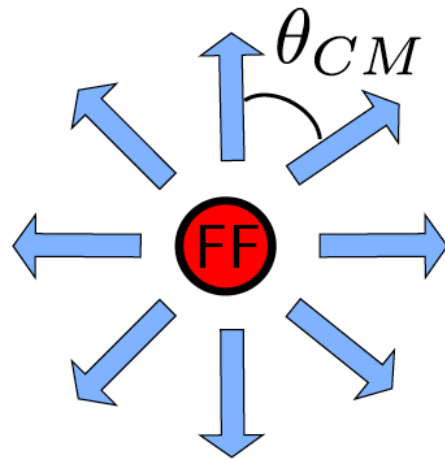
Boikov et al., EXFOR: 41110



A. Tudora et al. et al., ANE 35 (2008) 1131

How to measure neutrons

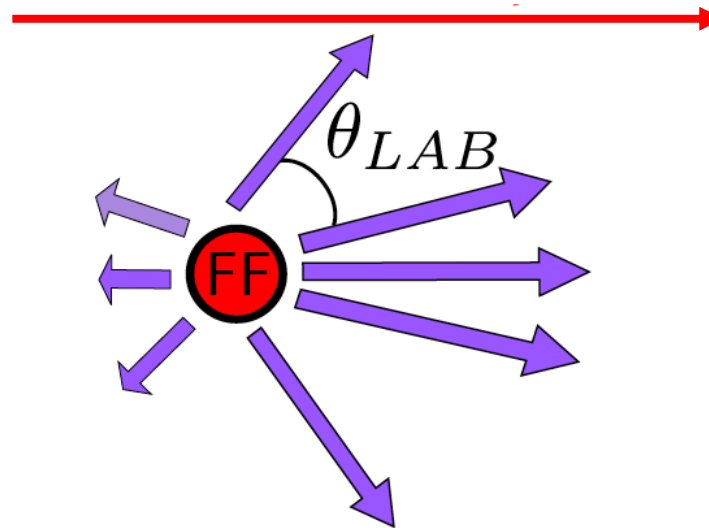
➤ Prompt neutron measurements



**center of mass
frame**

To extract physics

fragment velocity

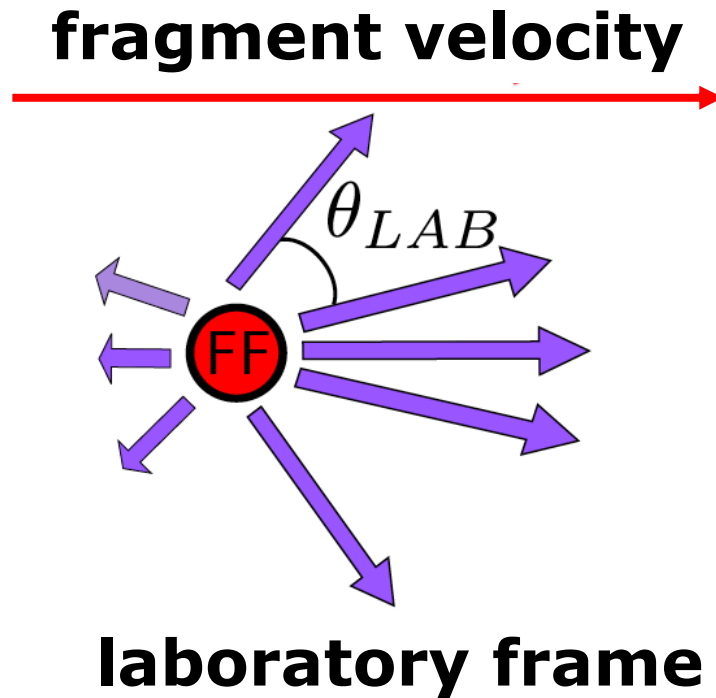
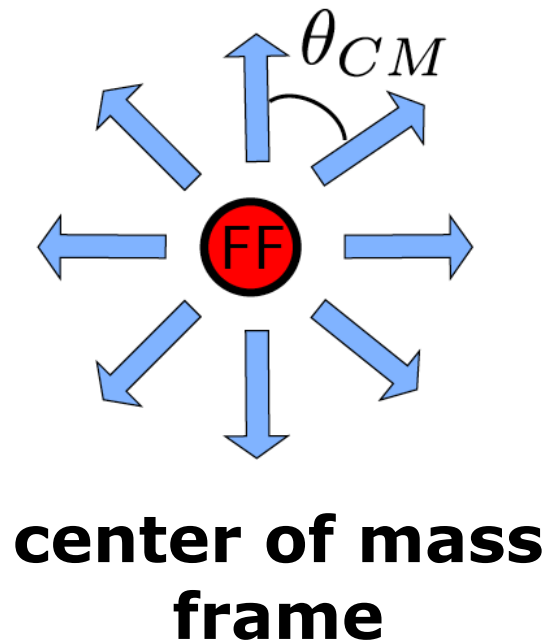


laboratory frame

**What you measure,
relevant for application**

How to measure neutrons

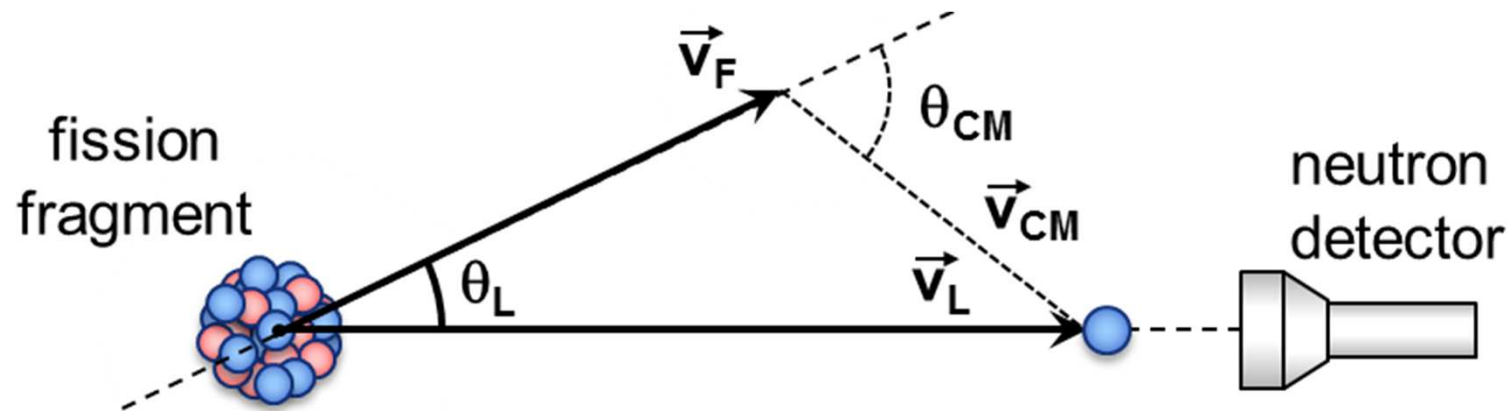
- And for extracting physics, remember this:



Transformation from the LS to CMS

How to measure neutrons

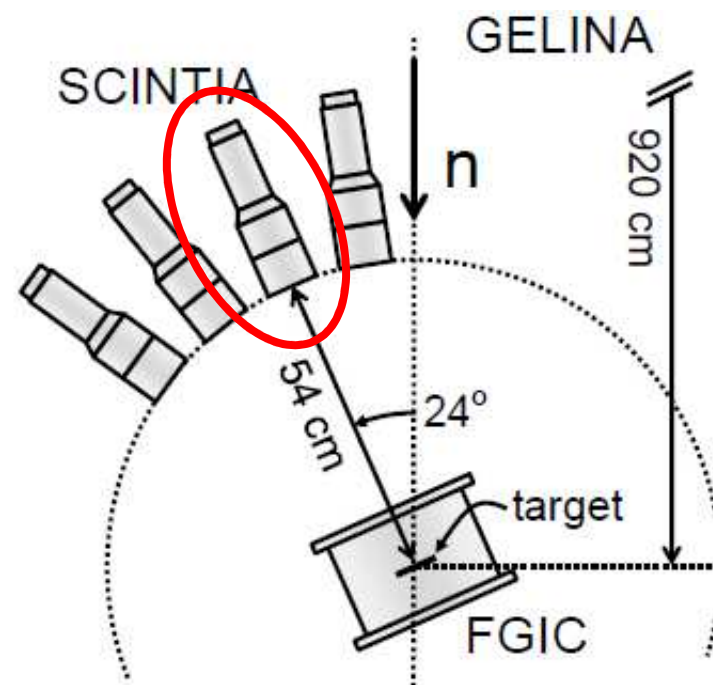
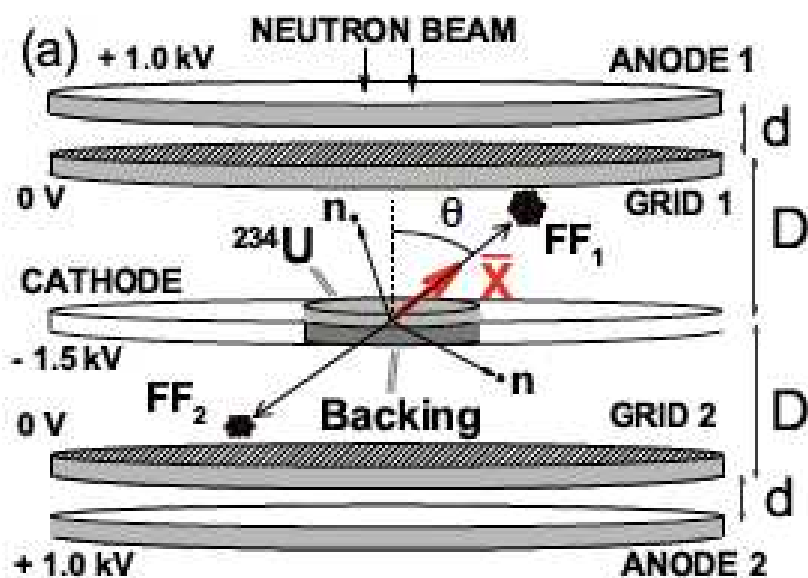
- **Emission of neutrons from fully accelerated fragments**
 - Obtain basic **kinematic information** in laboratory-frame
 - **Reconstruct emission process** in fission fragment rest-frame



- **Unbiased selection of events: $\cos\theta_{CM} \geq 0$**

How to measure neutrons

- The angle between the fission axis and the neutron vector is needed:



How to measure neutrons

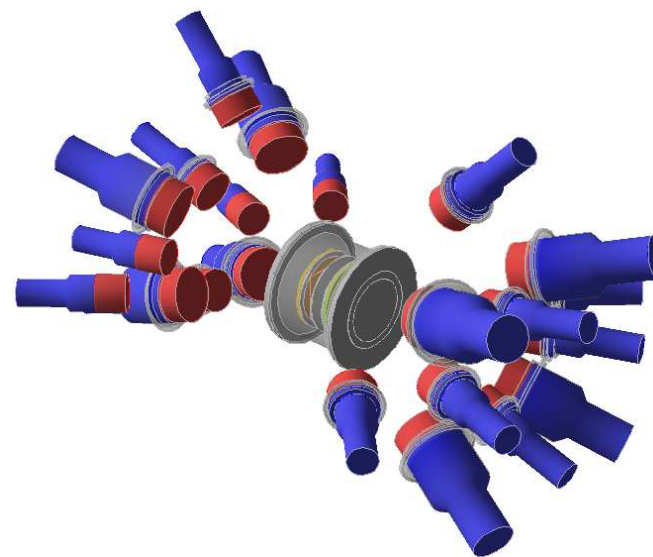


➤ **SCINTIA array (JRC-Geel)**

✓ **Array of 22 neutron detectors**

- ✓ SCIONIX LS301 (different sizes)
- ✓ P-Therphenyl ($\Phi=8.5$ cm, $h=6.8$ cm)

✓ **Double Frisch-grid (θ, ϕ) - sensitive IC**



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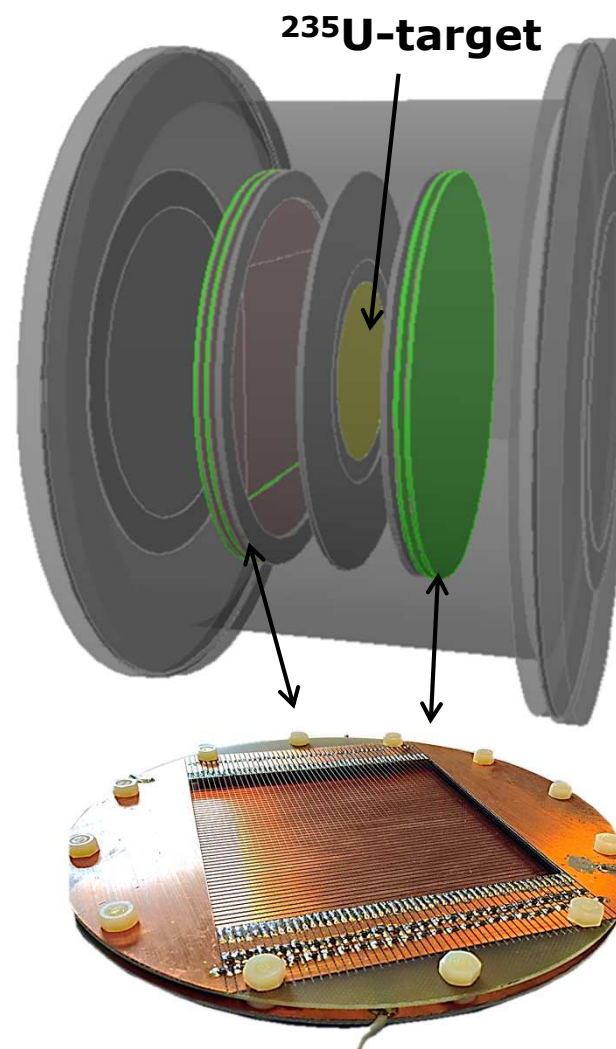
Fission fragment detector

Twin Ionization Chamber

- ✓ **Energies and Masses of fission fragments**
- ✓ **Large Geometrical Efficiency**
- ✓ **Timing resolution ~ 1 ns (FWHM)**
- ✓ **Polar angle θ of fission axis relative to the chamber axis**

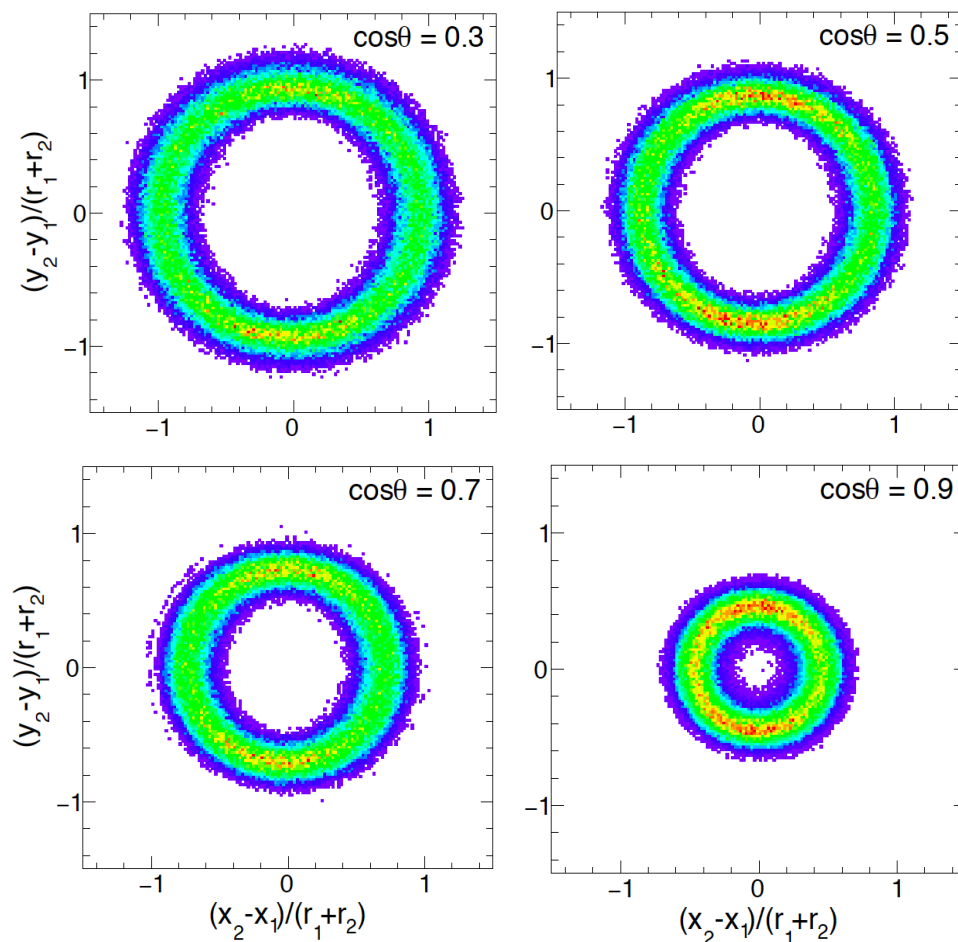
Position Sensitive Electrodes

- **Replaces anodes**
 - wire plane + strip anode
 - Projection of fission-axis on the electrode – plane
- ✓ **Fission axis orientation in 3D**



How to measure neutrons

Azimuthal angle of fission axis around chamber symmetry axis is determined from charge division read-out of position anodes



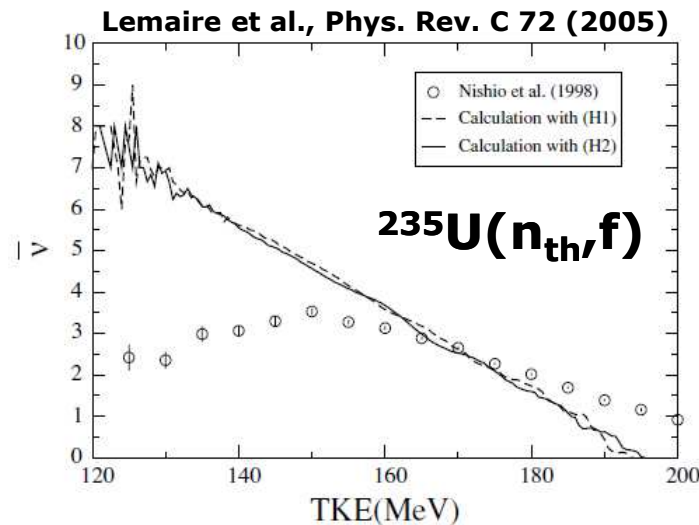
Difference in x-coordinates and y-coordinates of the Bragg-peak for the fission fragments detected on the opposite chamber sides.



A. Gök et al., NIMA 830 (2016) 366

Motivation

PFN multiplicity correlations with fragment observables

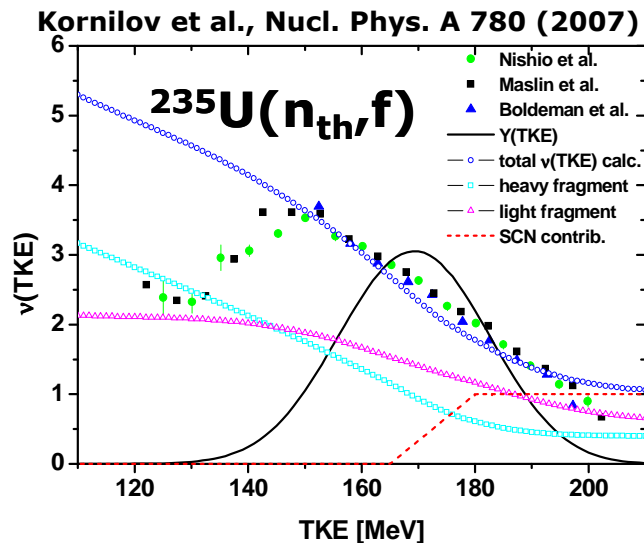


Based on energy balance in fission

- Detailed modelling (CGMF, Fifielin, Freya...)
 - successfully reproducing correlations
 - in the case $^{235}\text{U}(n, f)$
 - » difficulties: in particular $\nu(\text{TKE})$

Lemaire et al. (2005)

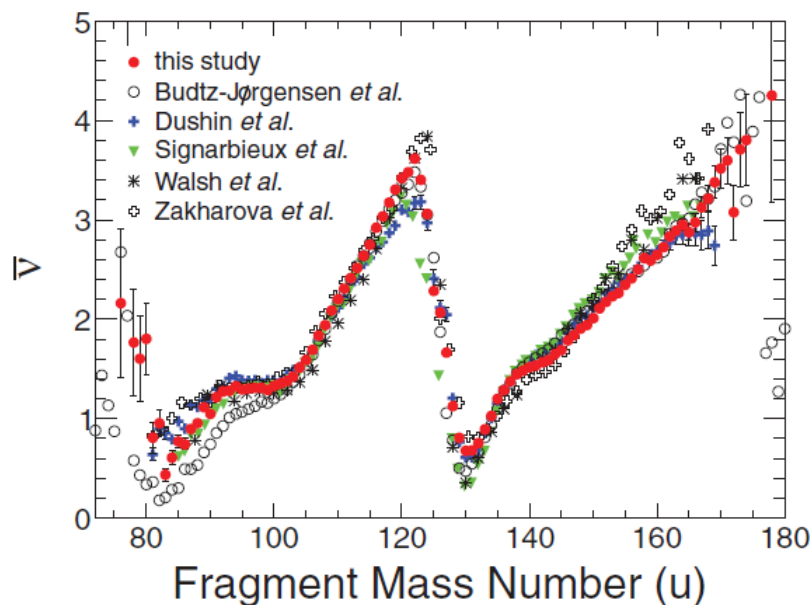
"...a dramatic deviation between calculation and experiment on ν is observed at low TKE that would indicate the presence of additional opened channels"



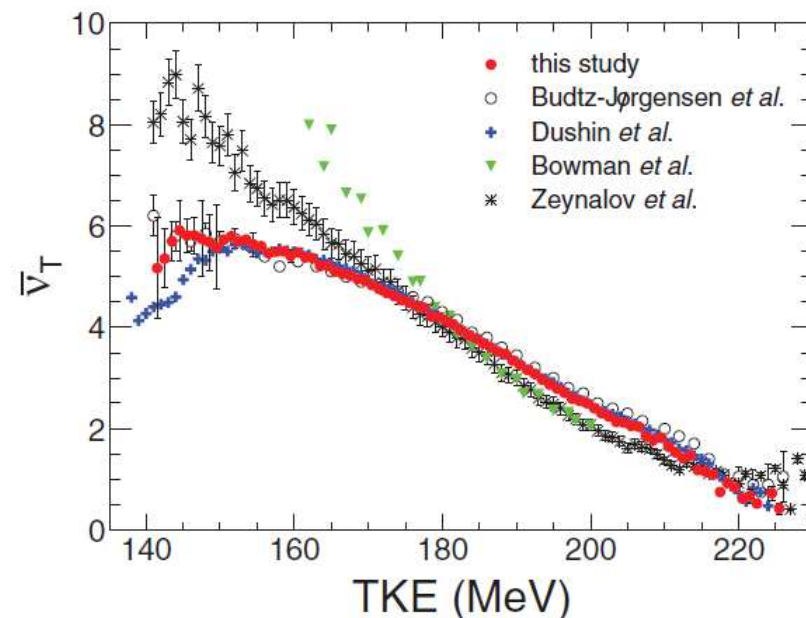
Kornilov et al. (2007)

"The incorporation of the SCN emission leads to a much better agreement between theoretical and experimental data for $\nu(\text{TKE})$ in the high energy range. However, the assumption of SCN emission at high TKE should be confirmed with direct experimental data"

Validation of method $^{252}\text{Cf}(\text{sf})$

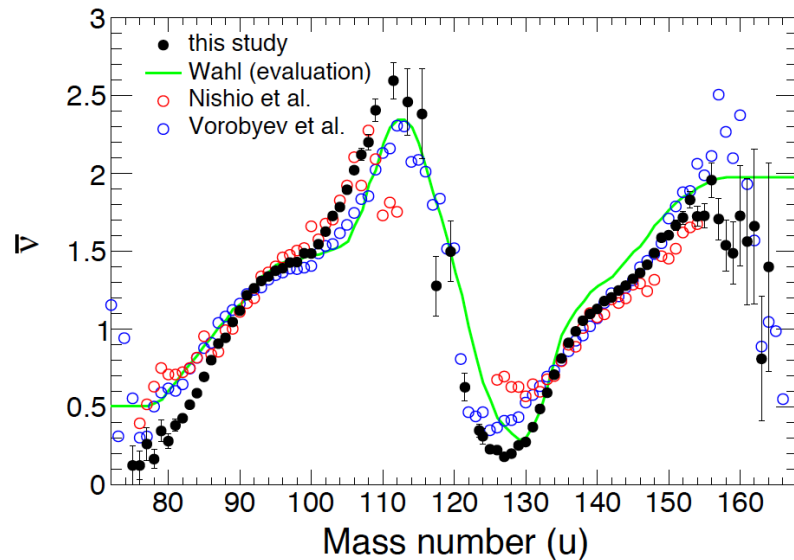


PHYSICAL REVIEW C **90**, 064611 (2014)



- Results show consistency with literature data
- Specifically with methods that do not suffer from neutron energy detection threshold
 - ✦ (Dushin *et al.*) Gd-loaded 4π scintillator tank

Multiplicity vs. Fragment Mass



Neutrons per fragment

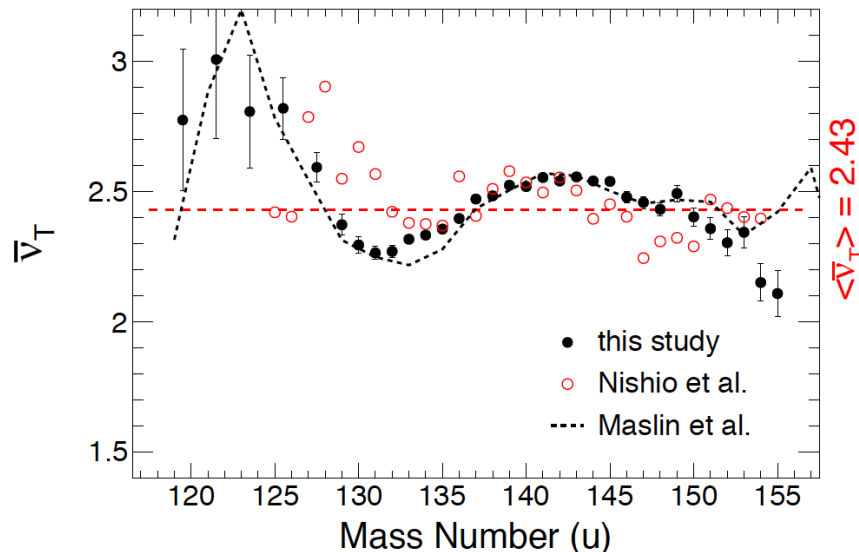
Saw-tooth distribution

Shoulders around

$$A_L=100 \text{ and } A_H=140$$

Pronounced minima around

$$A_L=80 \text{ and } A_H=130$$



Neutrons per fission

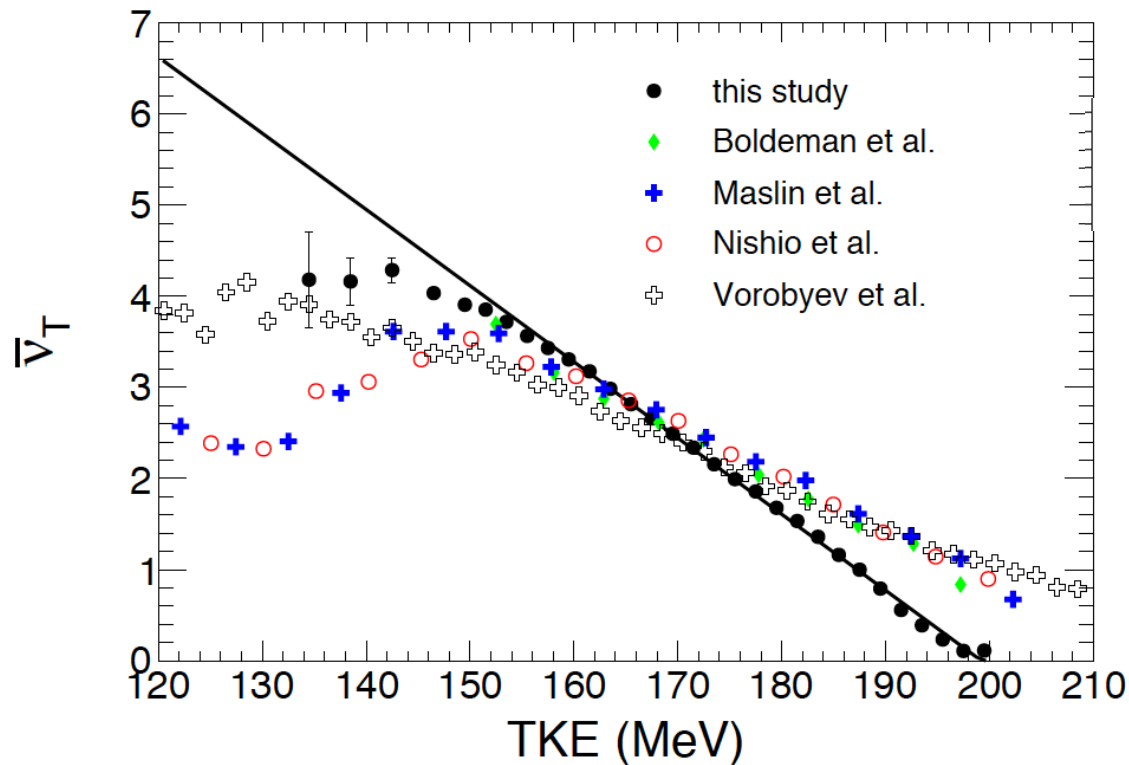
Flat distribution

Pronounced minimum around

$$A_H=132$$

$$E_n \in [0.3 \text{ eV}, 60 \text{ keV}]$$

Multiplicity vs. Fragment TKE



Close to linear dependence

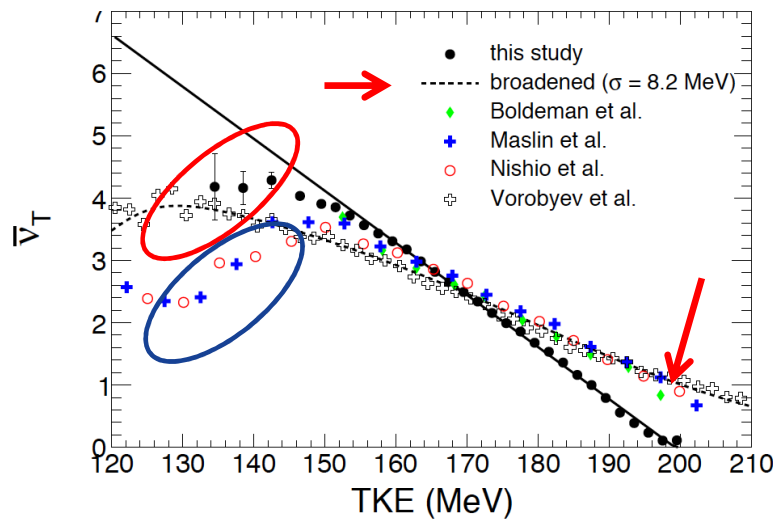
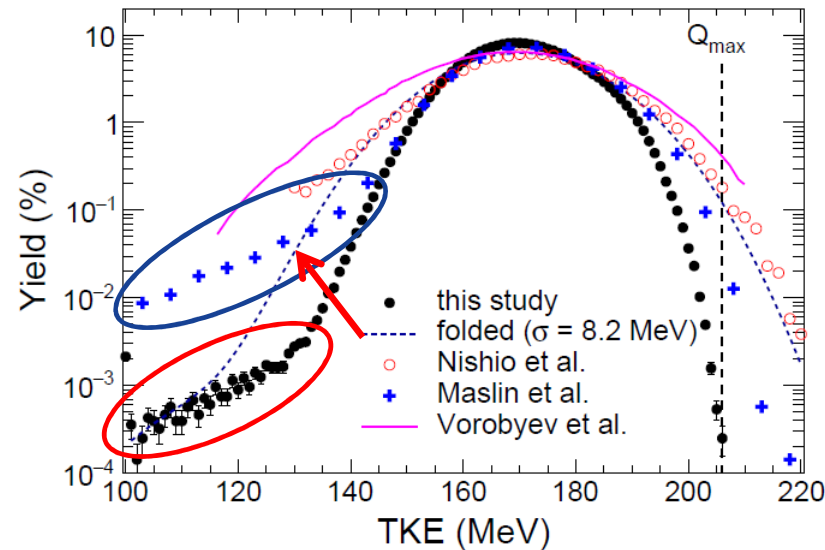
$$-\frac{dTKE}{d\bar{v}_T} = 12.0 \text{ MeV/n}$$

The slope is much steeper than earlier studies

$$-\frac{dTKE}{d\bar{v}_T} = 16.7 - 18.5 \text{ MeV/n}$$

The difference cannot be explained by difference in incident neutron energy

Multiplicity vs. Fragment TKE

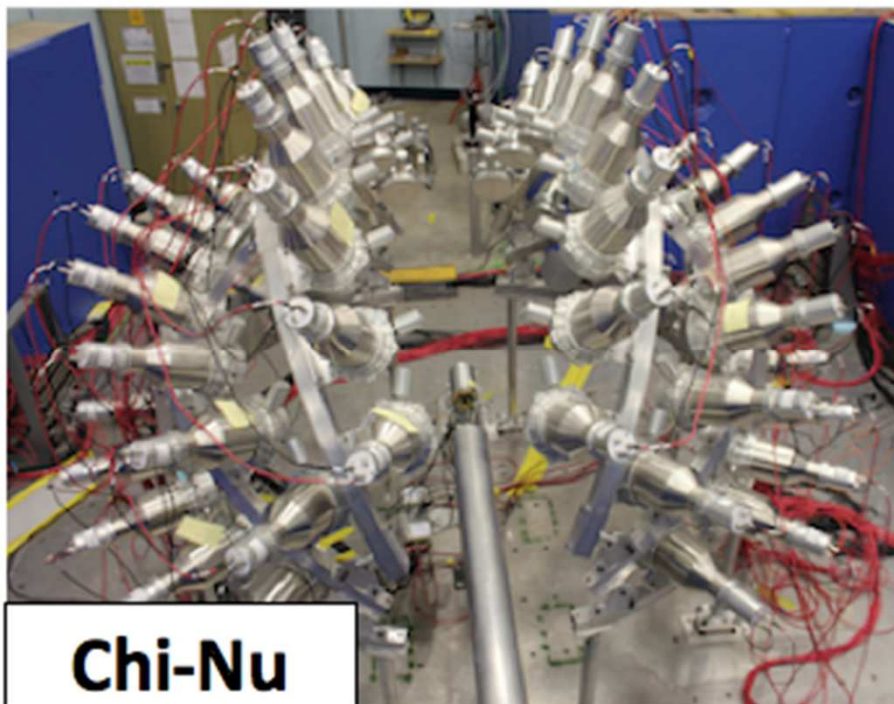


- Wide TKE-distributions
- Significant Yield at TKE > Q_{max}
- ⇒ Resolution broadening
- Decreased slope
- Increased neutron yield at Q_{max}

Tailing of TKE distribution

- Energy degraded scattered fission fragments
- Neutron yield should approach average nubar
- Drop in nubar at low TKE
- Present also in our data

How to measure neutrons



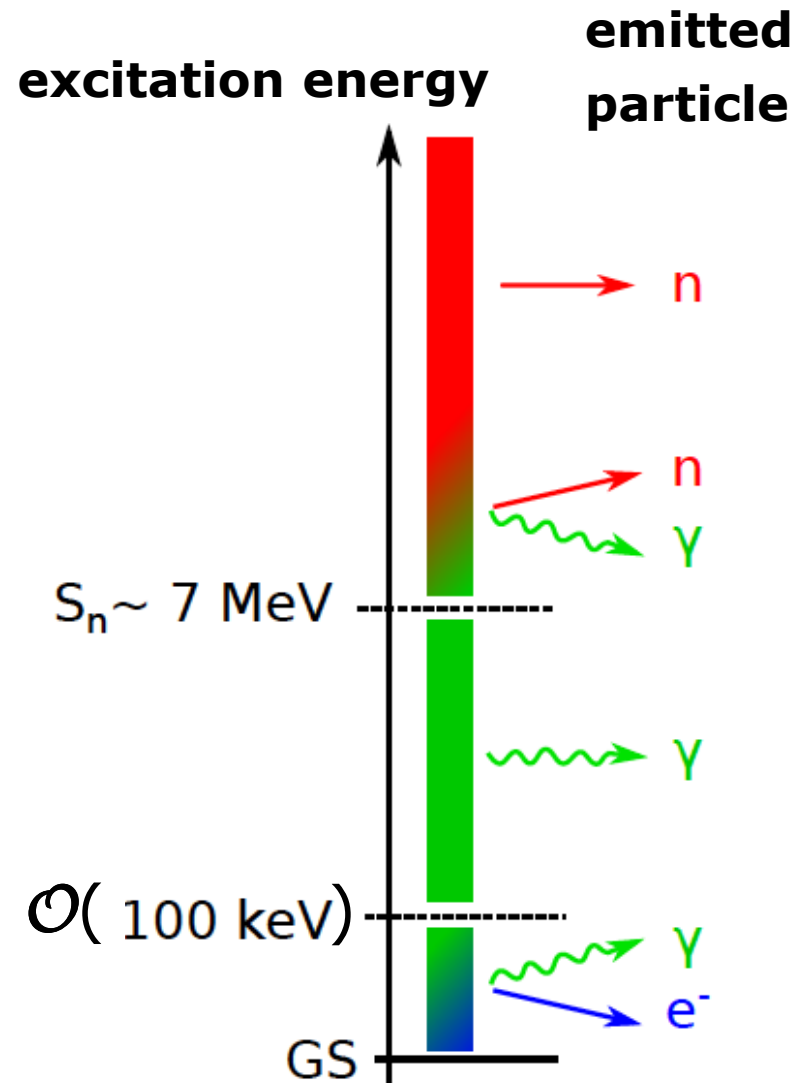
- ✓ Array of 54 neutron detectors
- ✓ Multi plate IC
- ✓ Measure PFNS as a fct of E_n

➤ Chi-nu array (LANL)

How to measure neutrons and γ -rays

➤ Prompt γ -ray measurements

How to measure prompt fission γ -rays



How to measure prompt fission γ -rays

➤ Separation of γ -rays from prompt neutrons

- Time-of-flight method
- Excellent timing resolution of the combined γ -ray and fission detector
- Determines the geometrical efficiency of your instrument

➤ Best possible energy resolution

How to measure prompt fission γ -rays

- **Fission detectors: FGIC, Si-detectors, single-crystal diamond detectors ($\sigma_t < 100$ ps)**

- **Choice of suitable γ -ray detectors:**
 - **High purity germanium detectors**
 - ✓ **Excellent energy resolution, bad timing resolution**
 - **Fragments moving \rightarrow Doppler broadened γ -peak**
 - **Very neutron sensitive**

How to measure prompt fission γ -rays

➤ Choice of suitable γ -ray detectors:

- **High purity germanium detectors**
- ✓ Excellent energy resolution, bad timing resolution
- Fragments moving → Doppler broadened γ -peak
- ~~Very neutron sensitive~~
- **Scintillation detectors**
- Limited energy resolution, worse peak-to-total
- ✓ In general much better timing resolution
- ✓ Higher efficiency, larger sizes available

How to measure prompt fission γ -rays

➤ Scintillation detectors:

In the 1970s sodium iodine (NaI) was used

- ✓ Timing resolution of the order of 5 – 7 ns
- ✓ Energy resolution 7% @ 662 keV
- ✓ TOF distance 1m or larger
- Limited geometrical efficiency

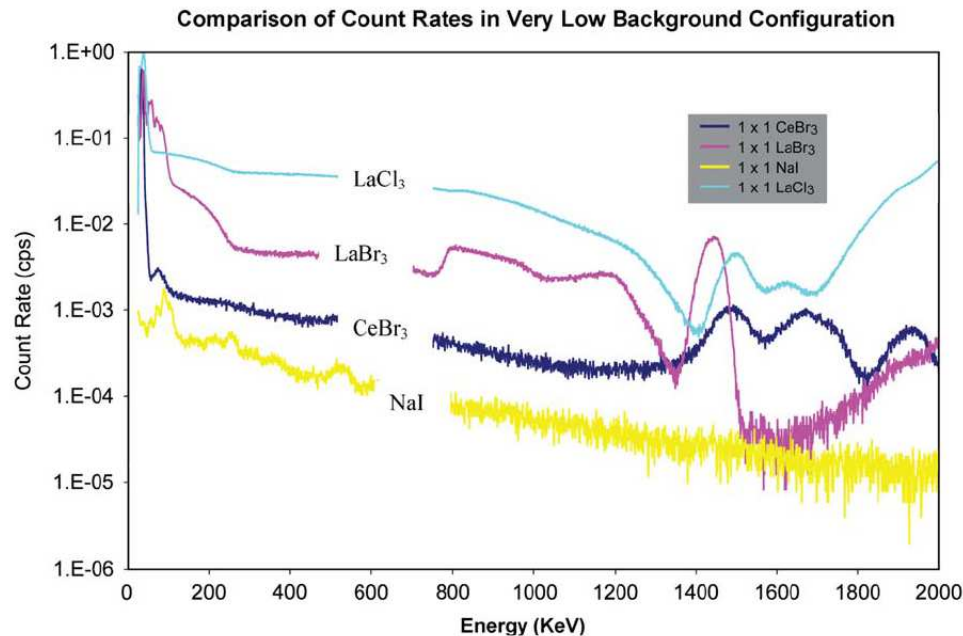
Nowadays we have to cope with limited resources in terms of beam time, staff...

Need of a more compact, efficient set-up!

How to measure prompt fission γ -rays

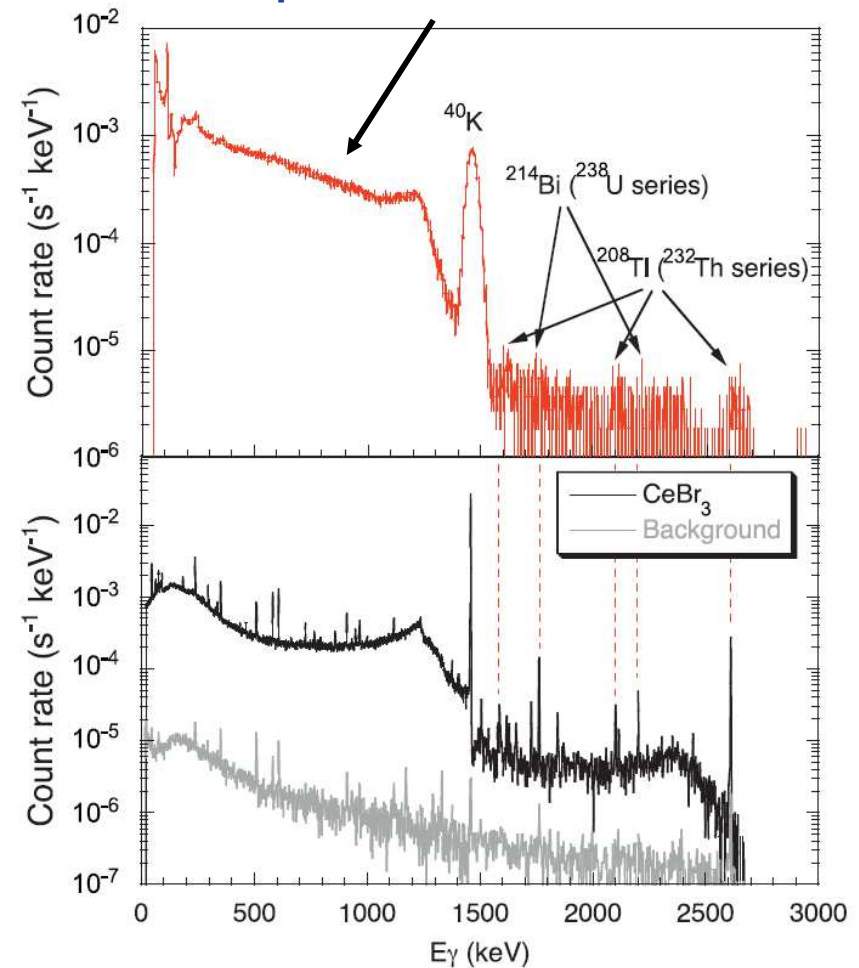
➤ Lanthanide halide detectors:

➤ Intrinsic activity



P. Guss et al. NIMA 608 (2009) 297

Compton scatter from PM tube



13 days measured in HADES

R. Billnert et al. NIMA 647 (2011) 94

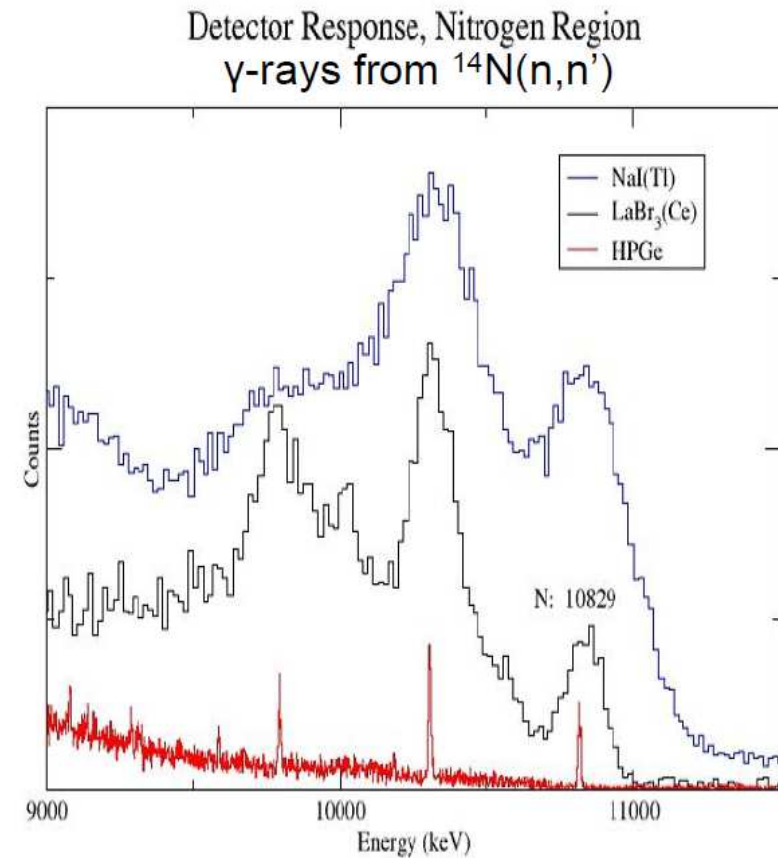
How to measure prompt fission γ -rays

➤ Lanthanide halide detectors:

➤ CeBr_3 detector



P. Guss et al. NIMA 608 (2009) 297

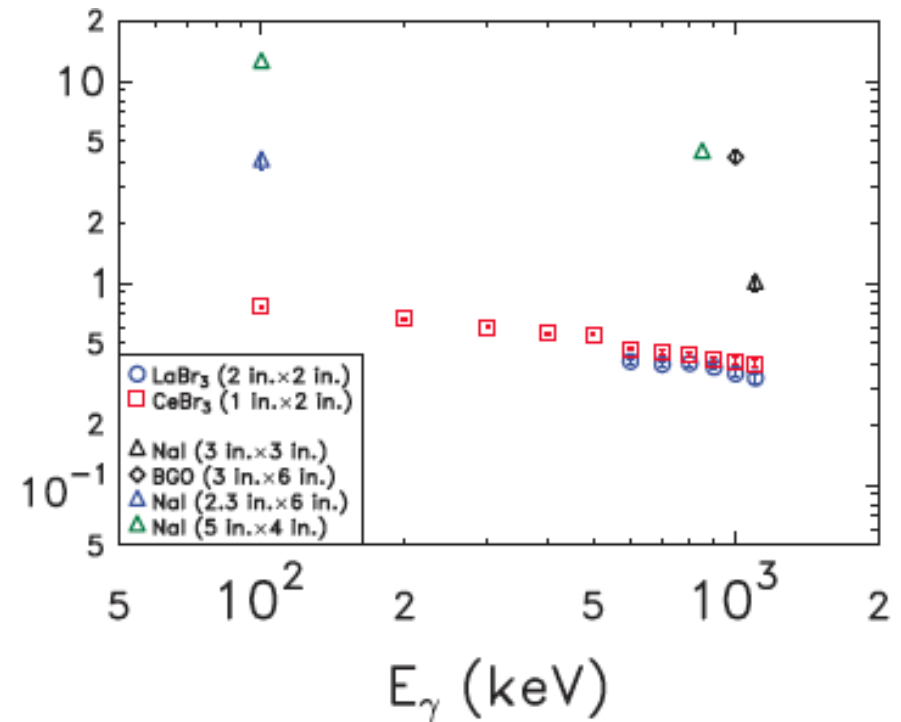
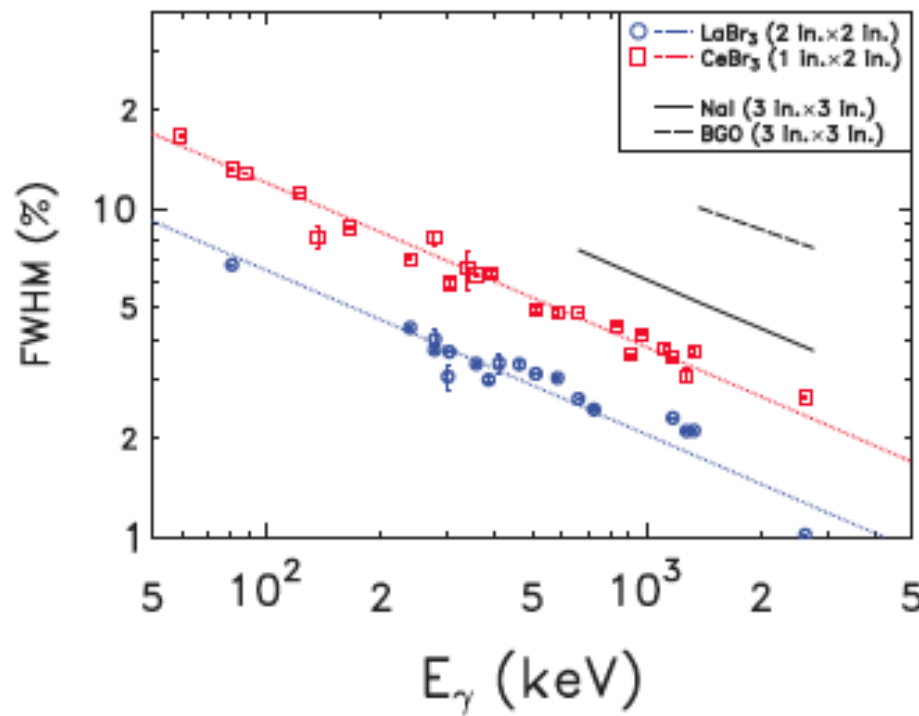


P. Guss, 2011 SPIE conf presentation

How to measure prompt fission γ -rays

➤ Lanthanide halide detectors:

- Cerium-doped lanthanum chloride ($\text{LaCl}_3:\text{Ce}$)
- Cerium-doped lanthanum bromide ($\text{LaBr}_3:\text{Ce}$)
- Cerium bromide (CeBr_3)
- (BGO: Bismuth Germanium Oxide)



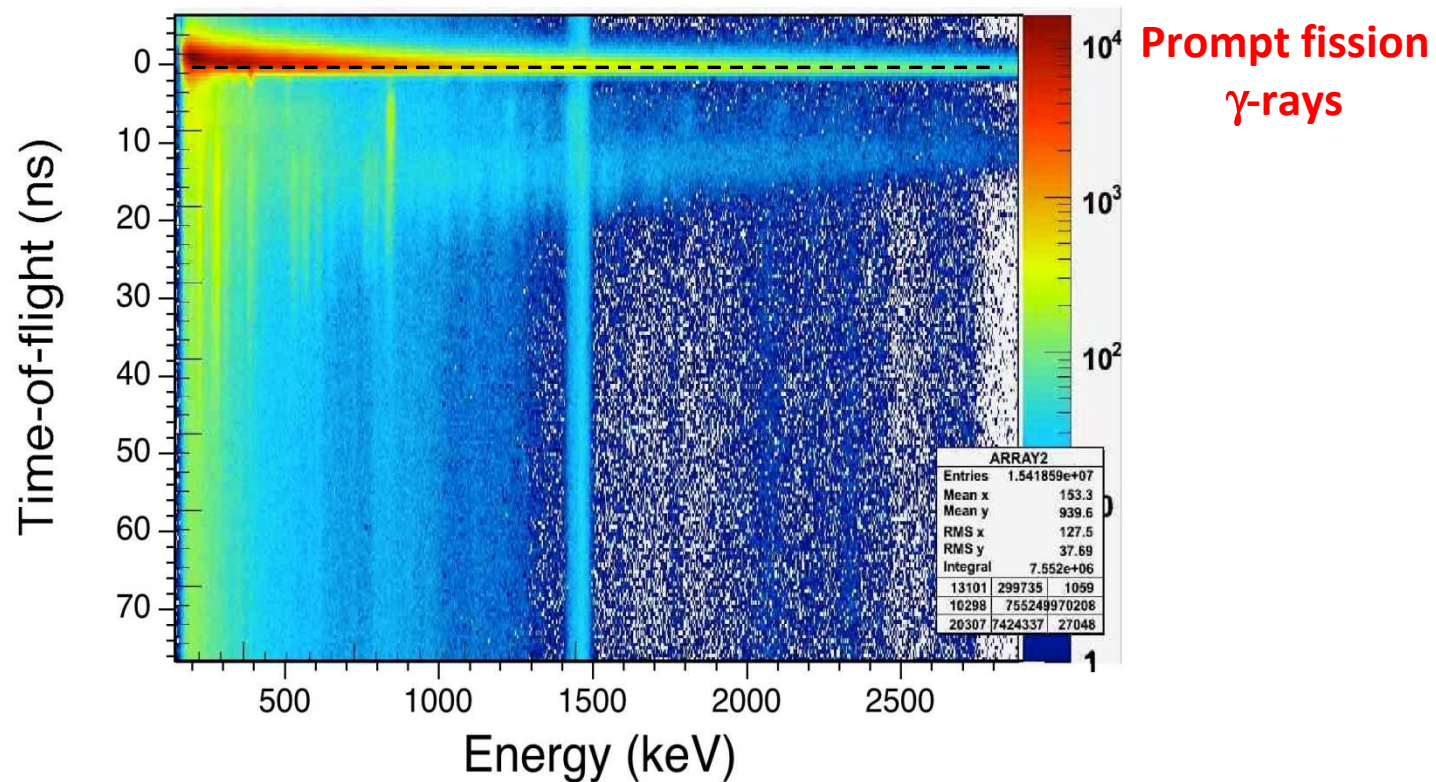
Energy resolution

Timing resolution
R. Billnert et al., PRC87 (2013) 024601



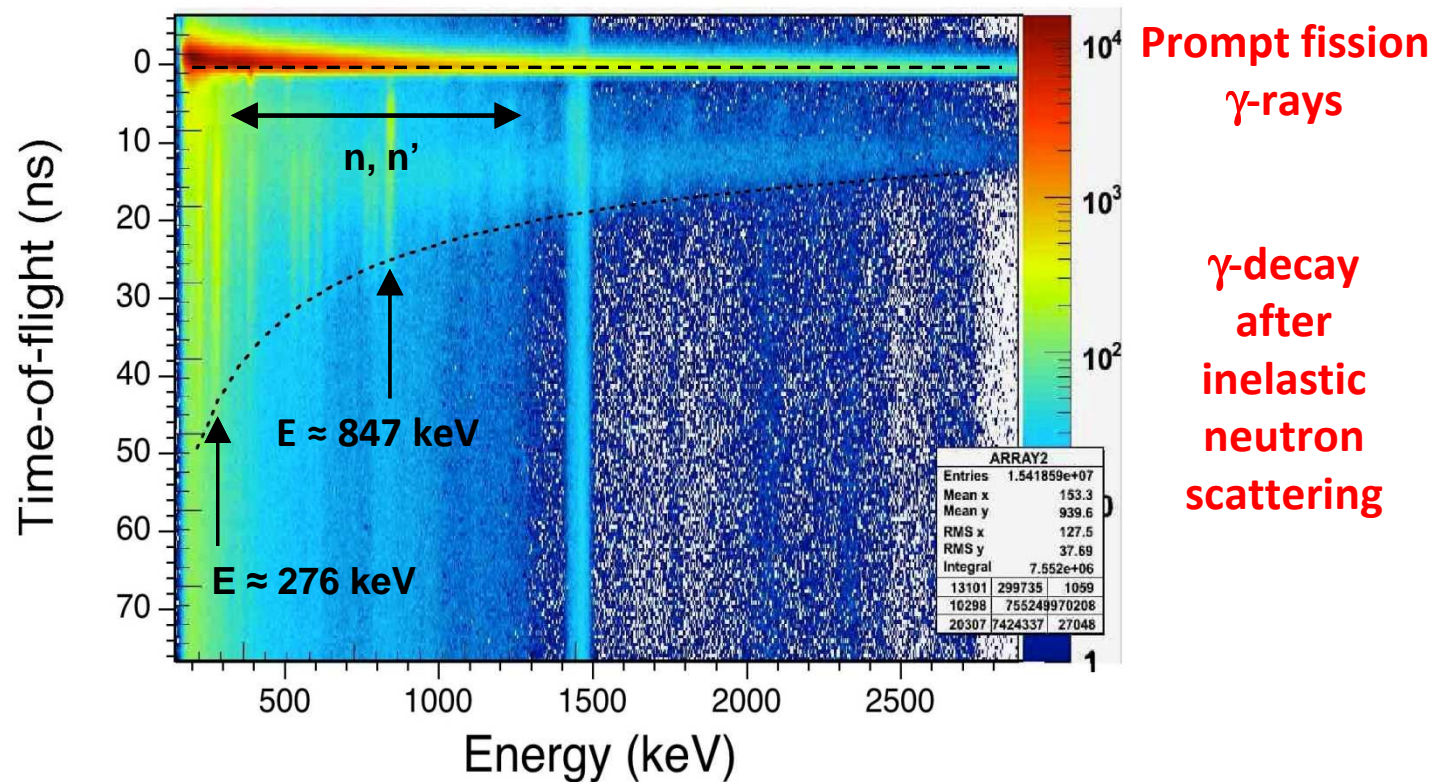
European
Commission

How to measure prompt fission γ -rays



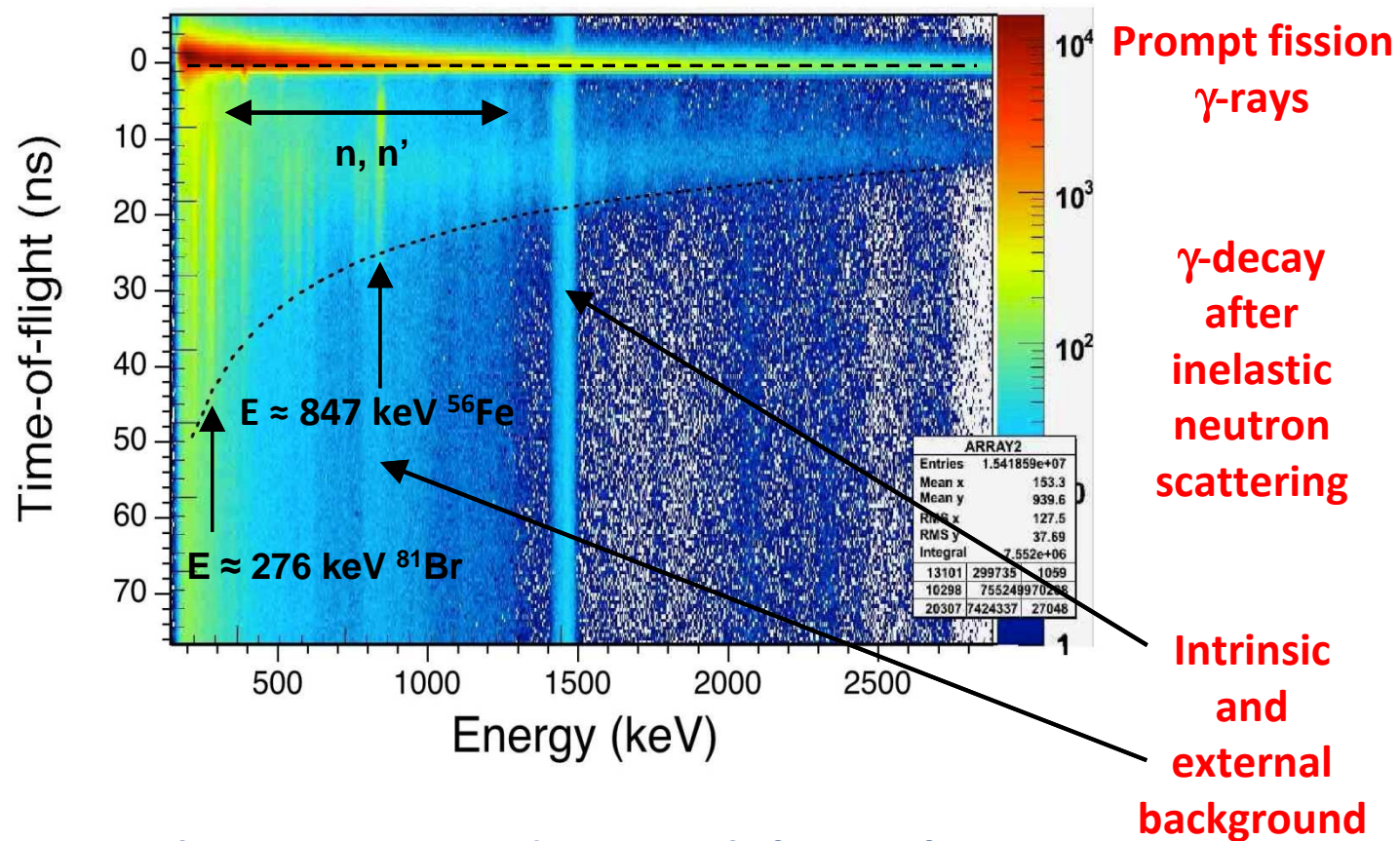
Photons in coincidence with fission fragments

How to measure prompt fission γ -rays



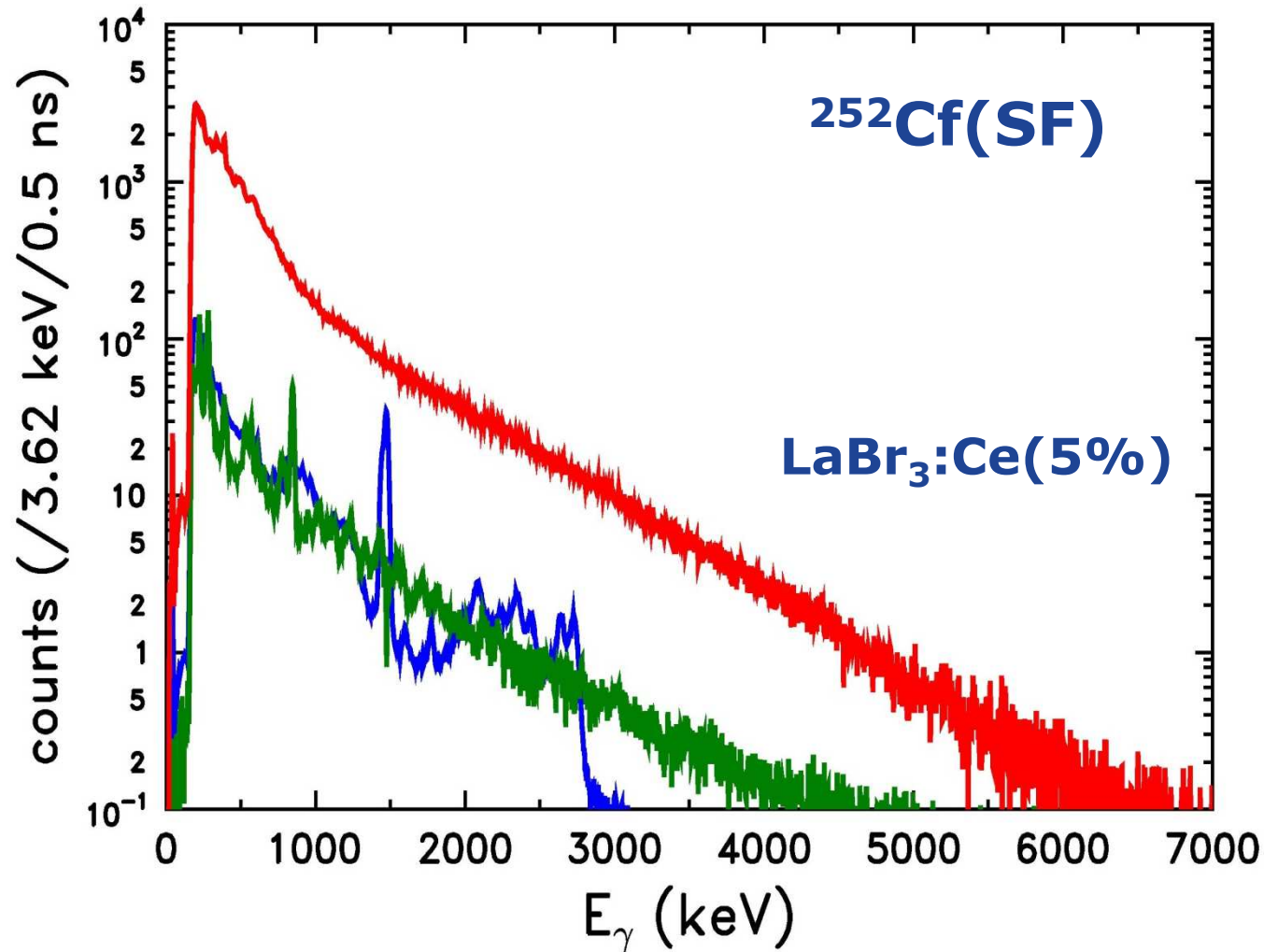
Photons in coincidence with fission fragments

How to measure prompt fission γ -rays



Photons in coincidence with fission fragments

How to measure prompt fission γ -rays



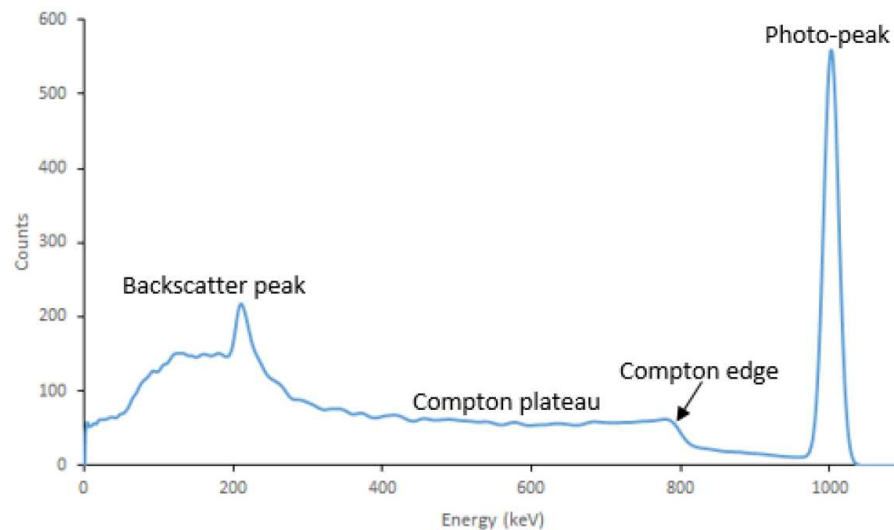
Red: PFGS, Green: Isomer decay, inelastic scattering, Blue: intrinsic bgrd



European
Commission |

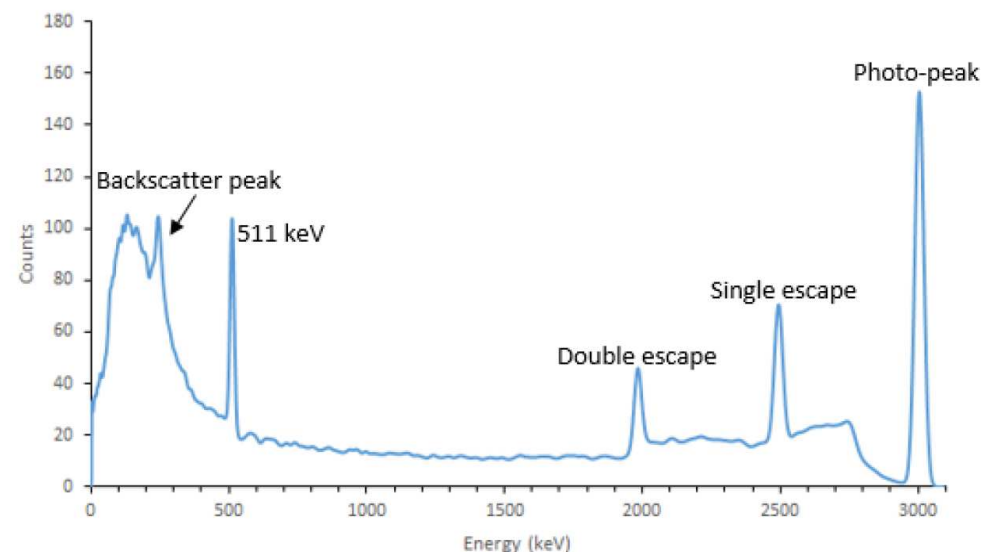
How to measure prompt fission γ -rays

➤ Detector response to different γ -ray energies



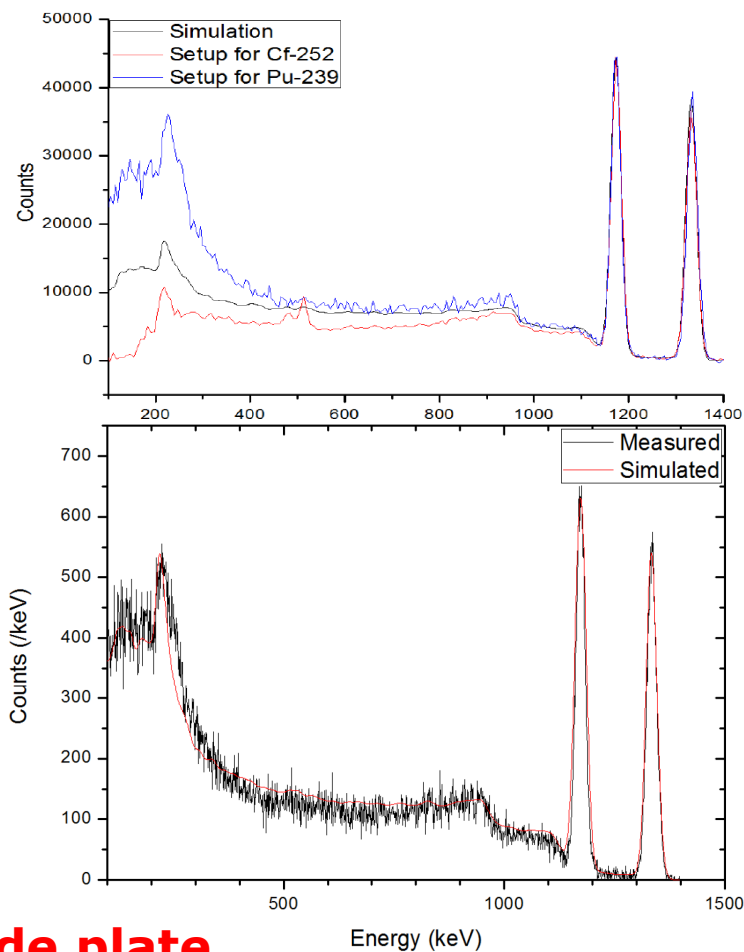
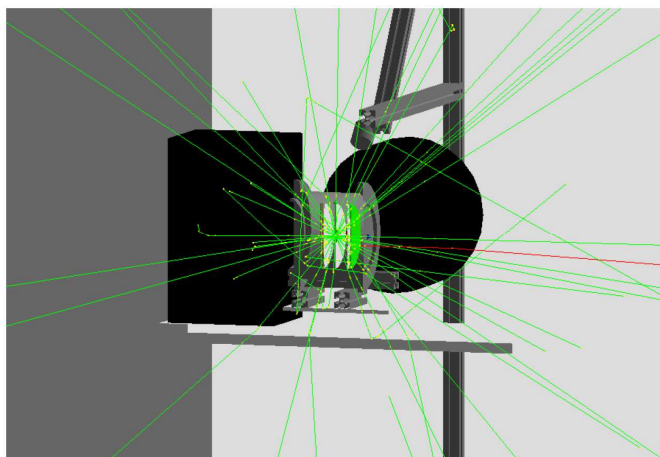
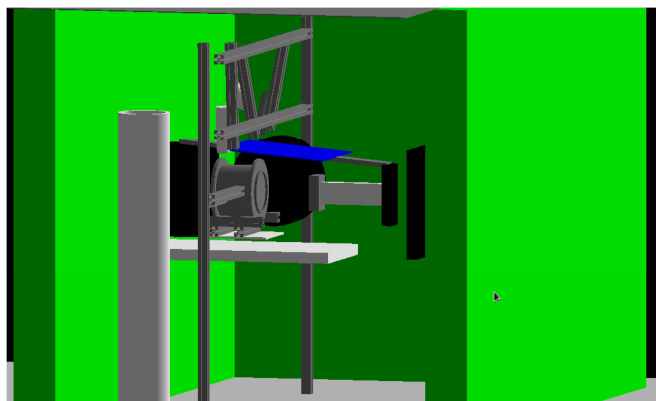
Simulated spectrum for 1 MeV photons hitting a 51 mm × 51 mm LaBr₃:Ce detector

Simulated spectrum for 3 MeV photons hitting a 51 mm × 51 mm LaBr₃:Ce detector



How to measure prompt fission γ -rays

- Optimized response function simulations to better reproduce energy range around the backscatter peak



Unexpected importance of cathode plate

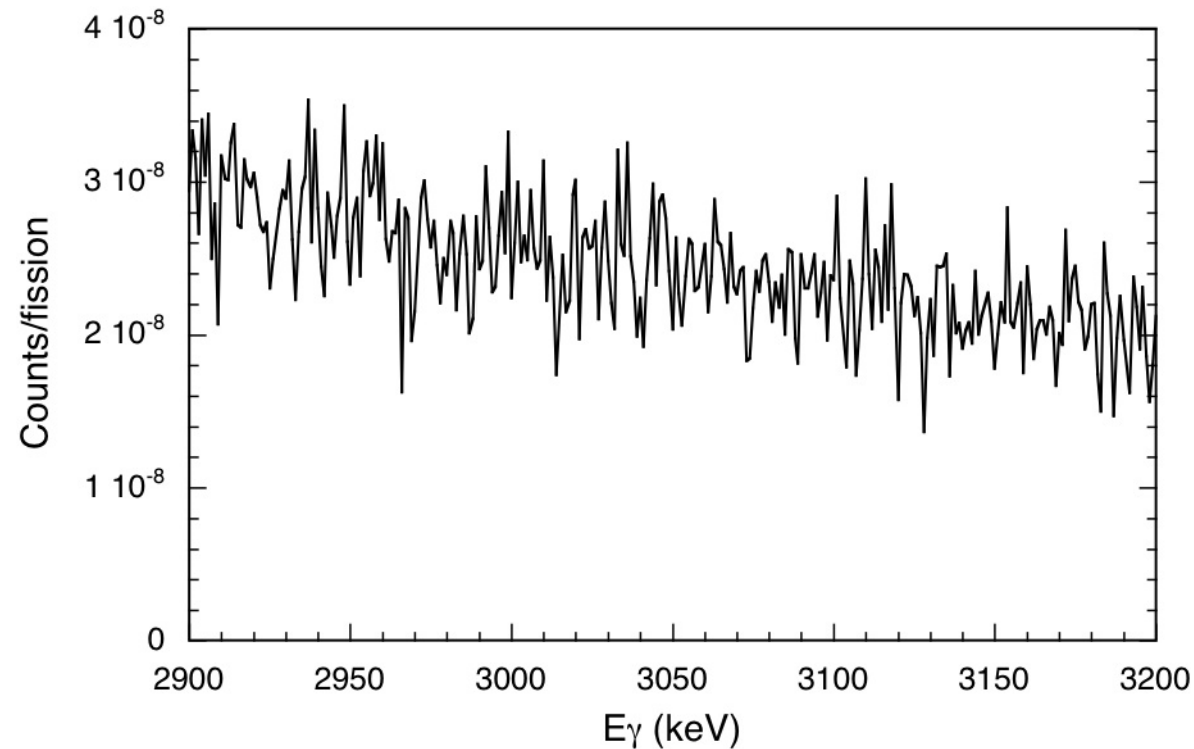
PhD thesis of Angélique Gatera

^{60}Co source



How to measure prompt fission γ -rays

➤ Unfolding the detector response

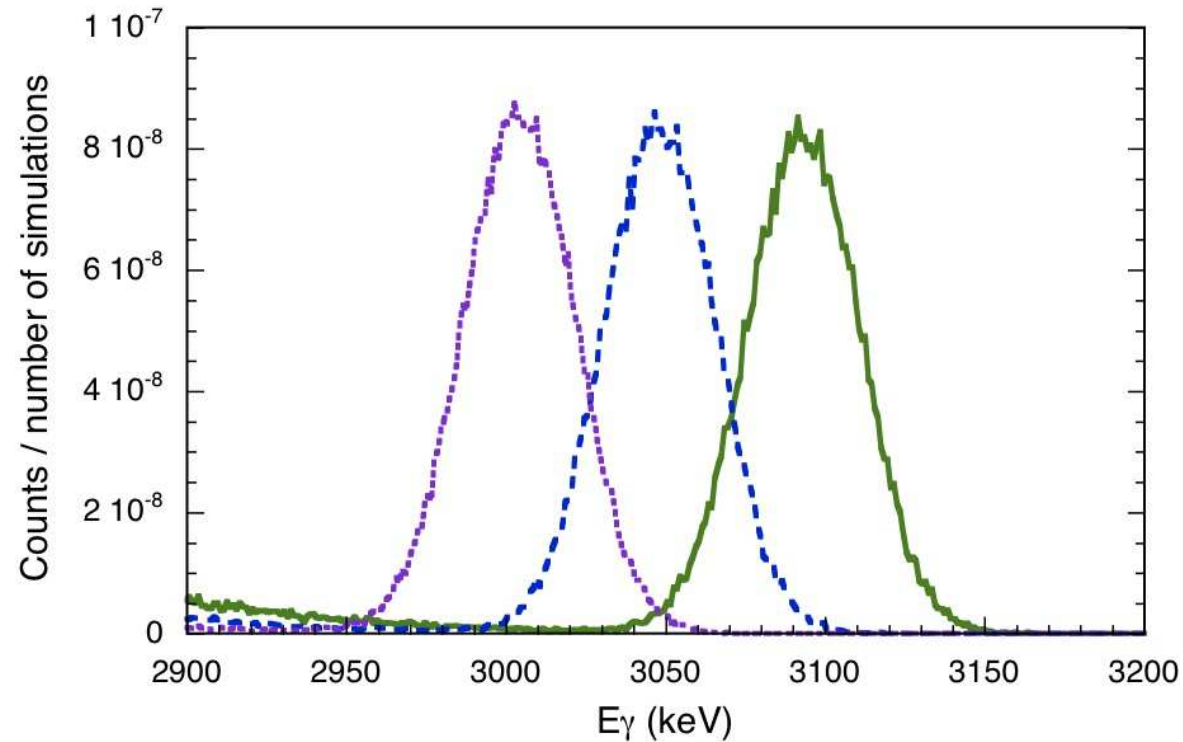


Measured $^{252}\text{Cf}(\text{SF})$ prompt fission γ -ray energy spectrum

→ e.g. zooming into region around 3 MeV

How to measure prompt fission γ -rays

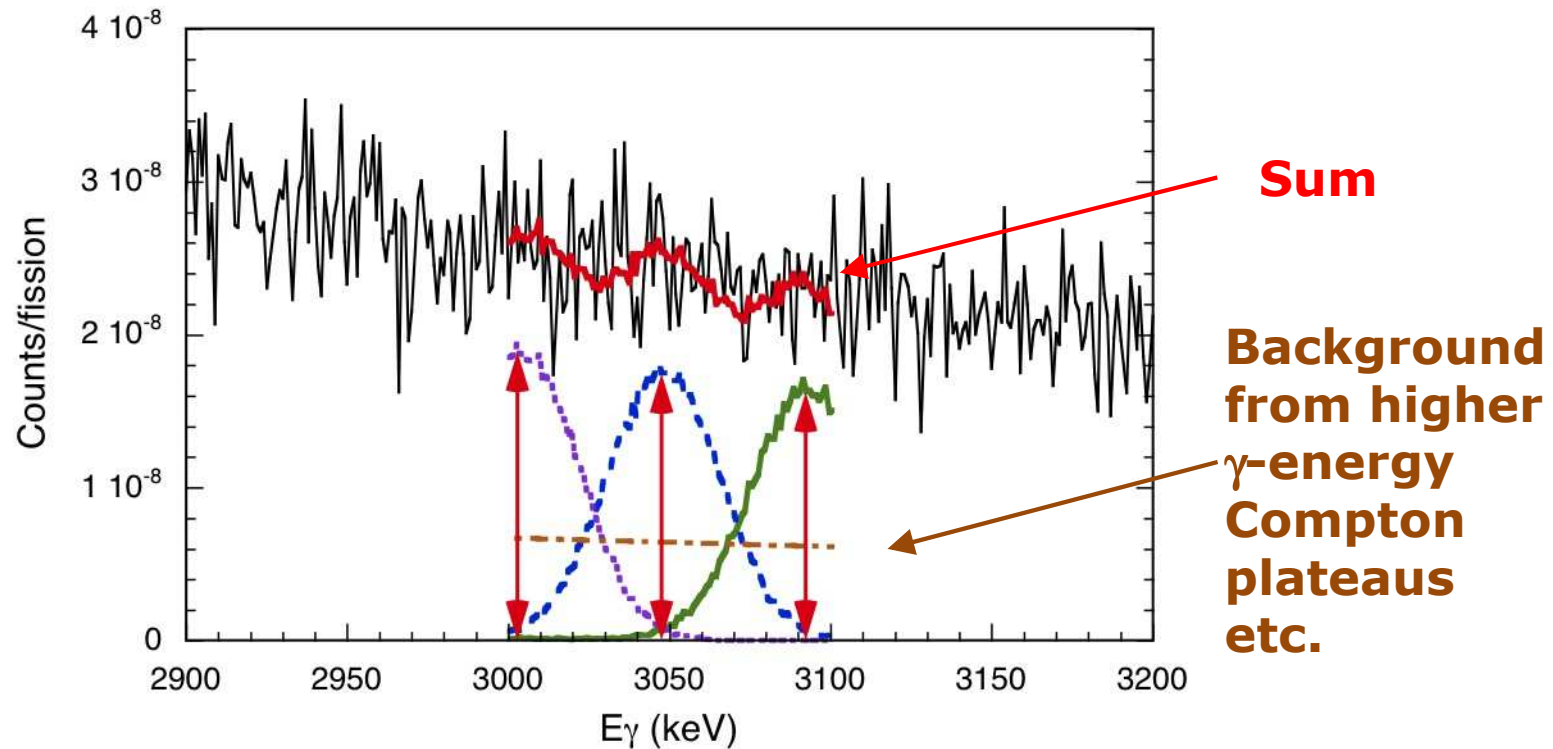
➤ Unfolding the detector response



**Simulating response function for mono-energetic γ -rays,
distance: FWHM from energy resolution measurements**

How to measure prompt fission γ -rays

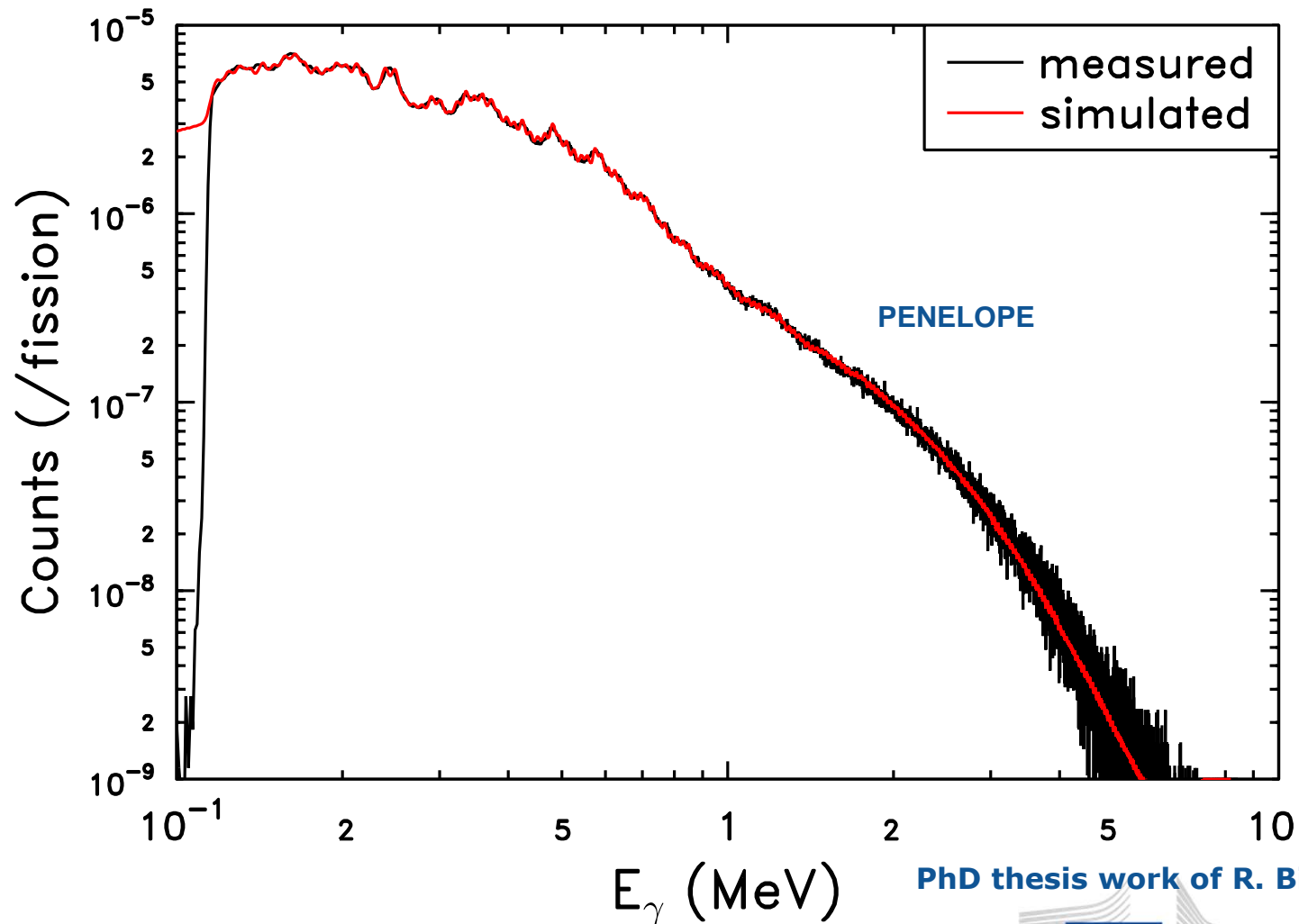
➤ Unfolding the detector response



Adjusting simulated spectra to measured γ -ray spectrum and determining the **scaling factors**

How to measure prompt fission γ -rays

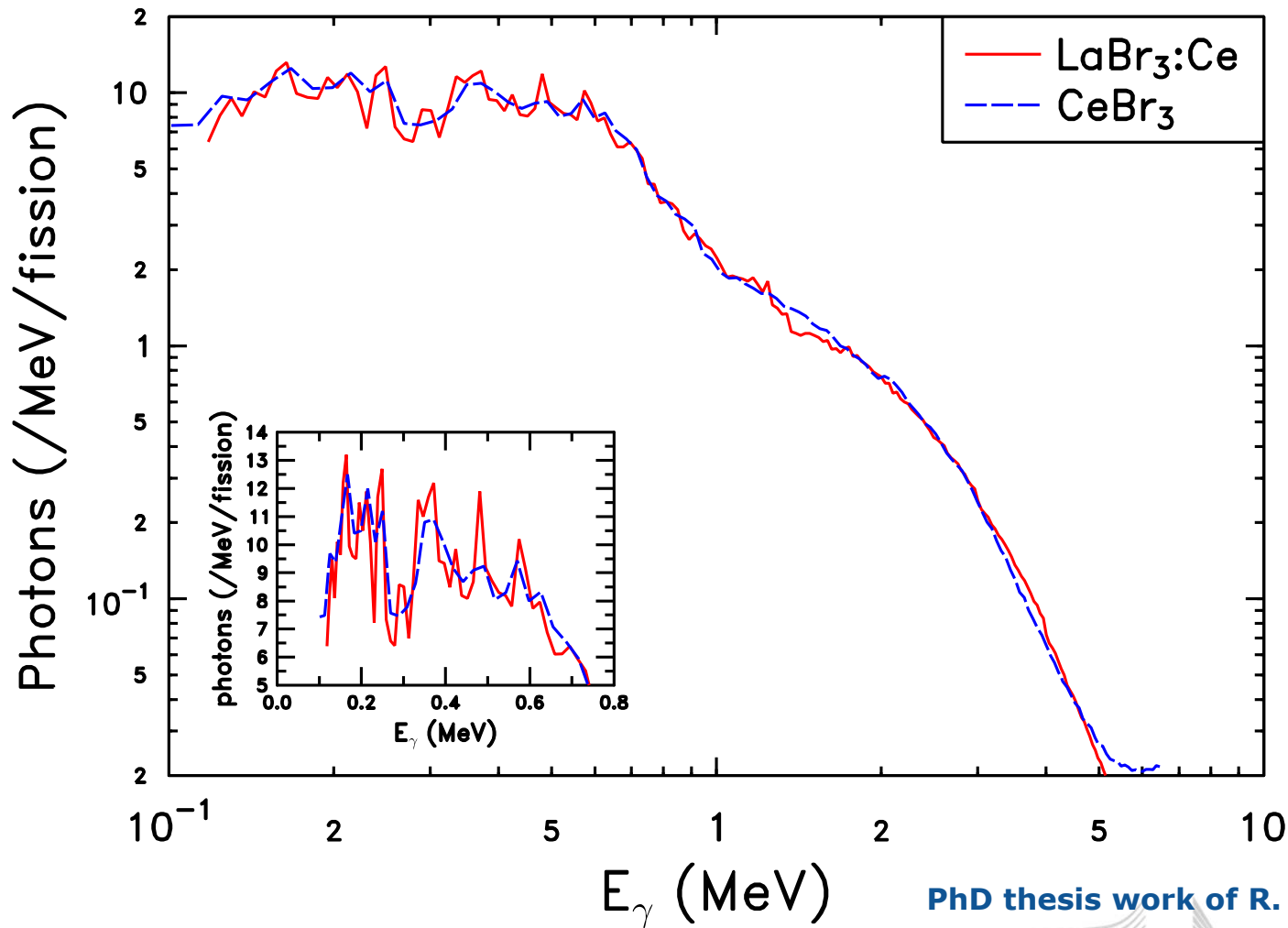
➤ Unfolding the detector response



PhD thesis work of R. Billnert

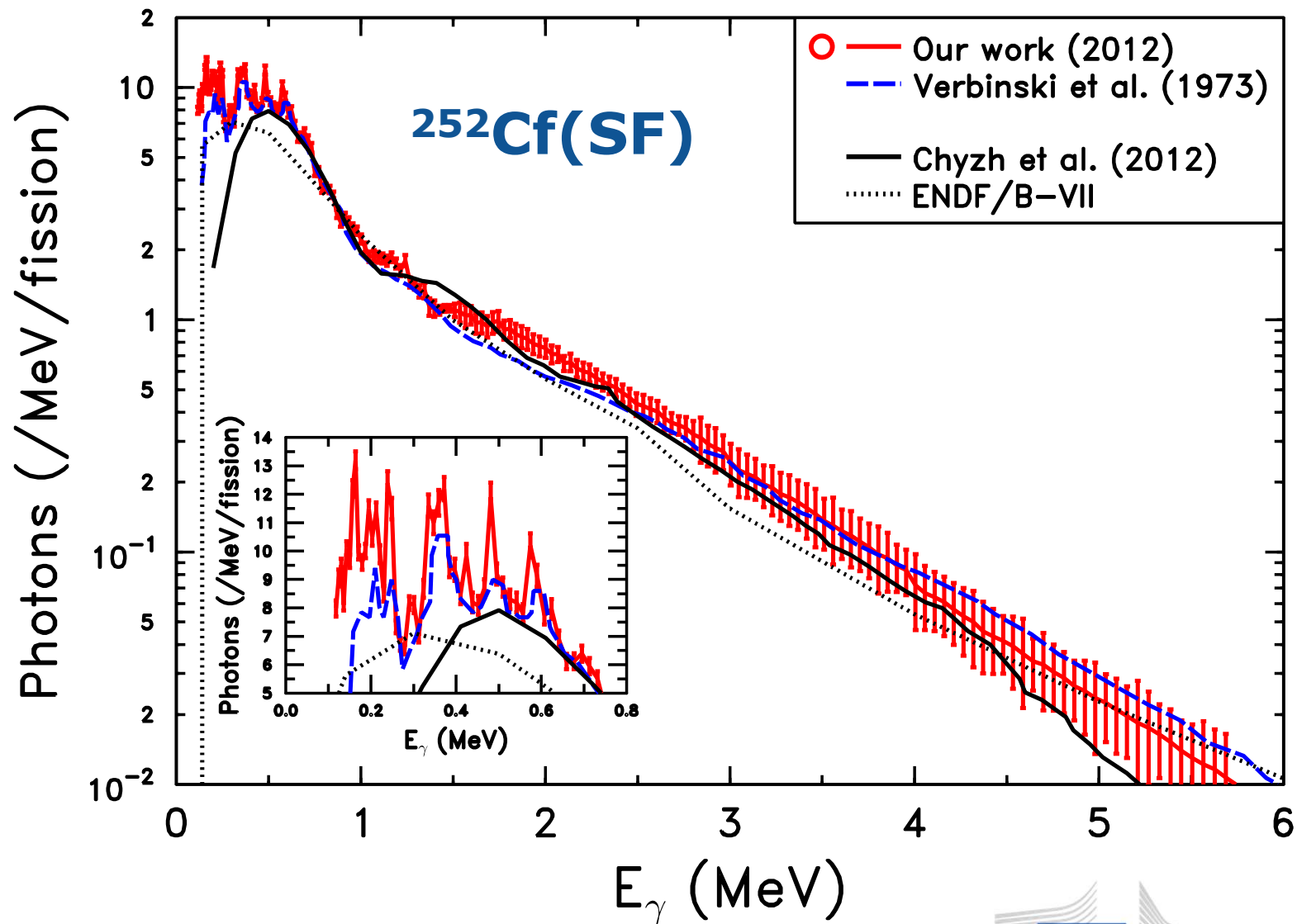
How to measure prompt fission γ -rays

➤ Unfolded emission spectrum

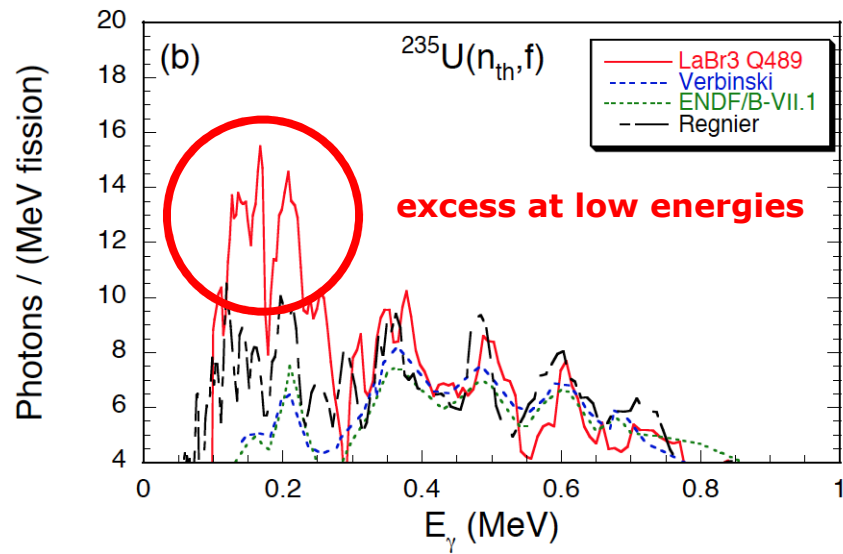


PhD thesis work of R. Billnert

How to measure prompt fission γ -rays



How to measure prompt fission γ -rays



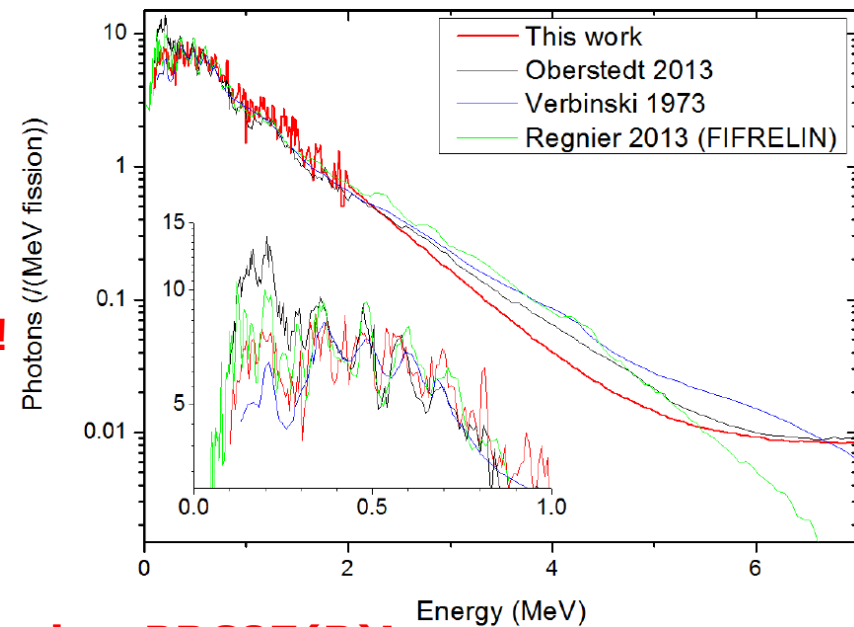
Situation as published in PRC87(R)

- $M_\gamma = (8.19 \pm 0.11) / \text{fission}$
- $E_{\gamma\text{tot}} = (6.92 \pm 0.09) \text{ MeV} / \text{fission}$

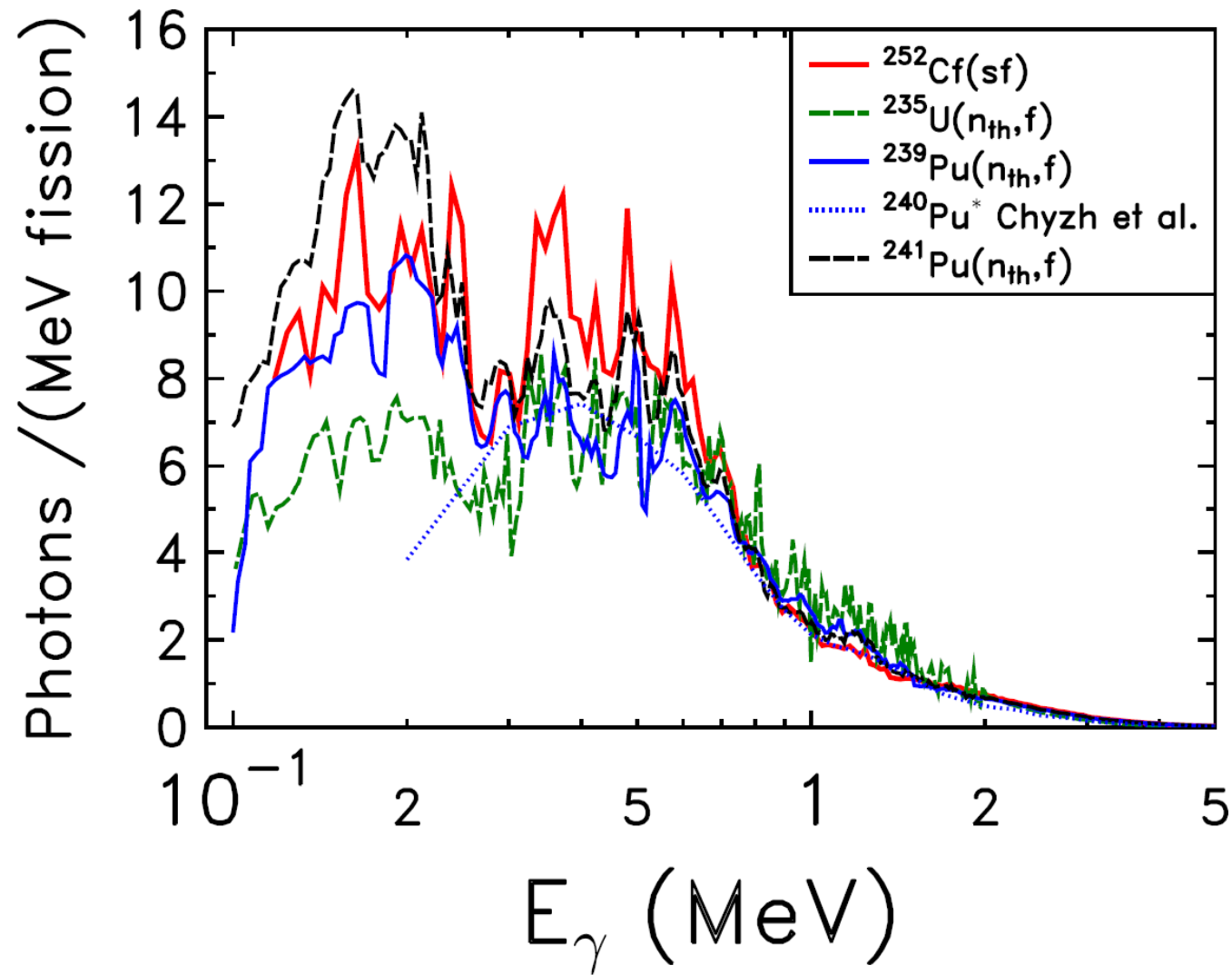
Situation after re-measuring
at the GELINA neutron TOF facility

New detector response calculations!

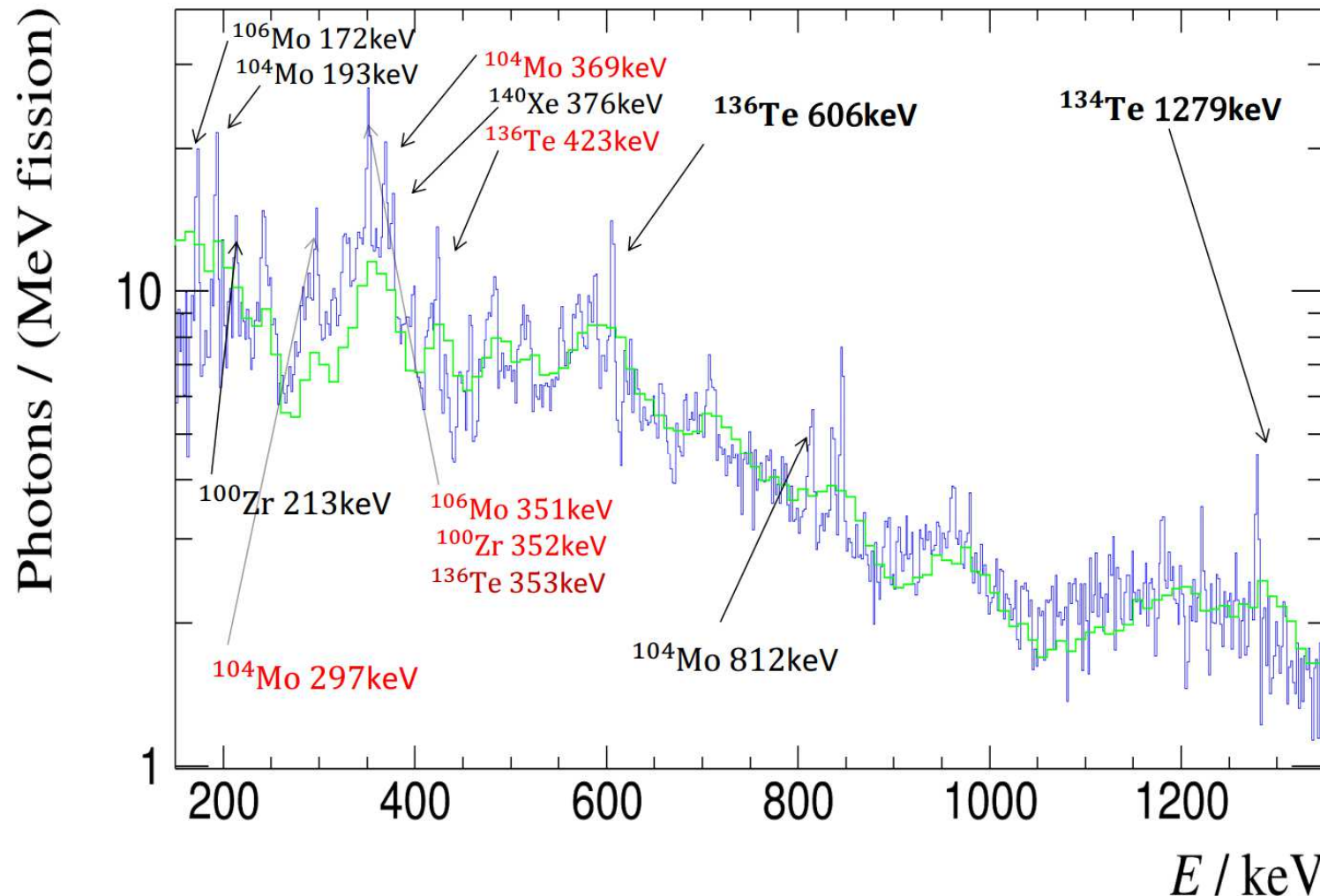
- ✓ $M_\gamma = (7.4 \pm 0.2) / \text{fission}$
- ✓ $E_{\gamma\text{tot}} = (6.4 \pm 0.2) \text{ MeV} / \text{fission}$
- ✓ Decrease of 10% and 8% compared to PRC87(R)!



How to measure prompt fission γ -rays



How to measure prompt fission γ -rays



$2+ \rightarrow 0+$ and $4+ \rightarrow 2+$ transitions of abundant FF are resolved

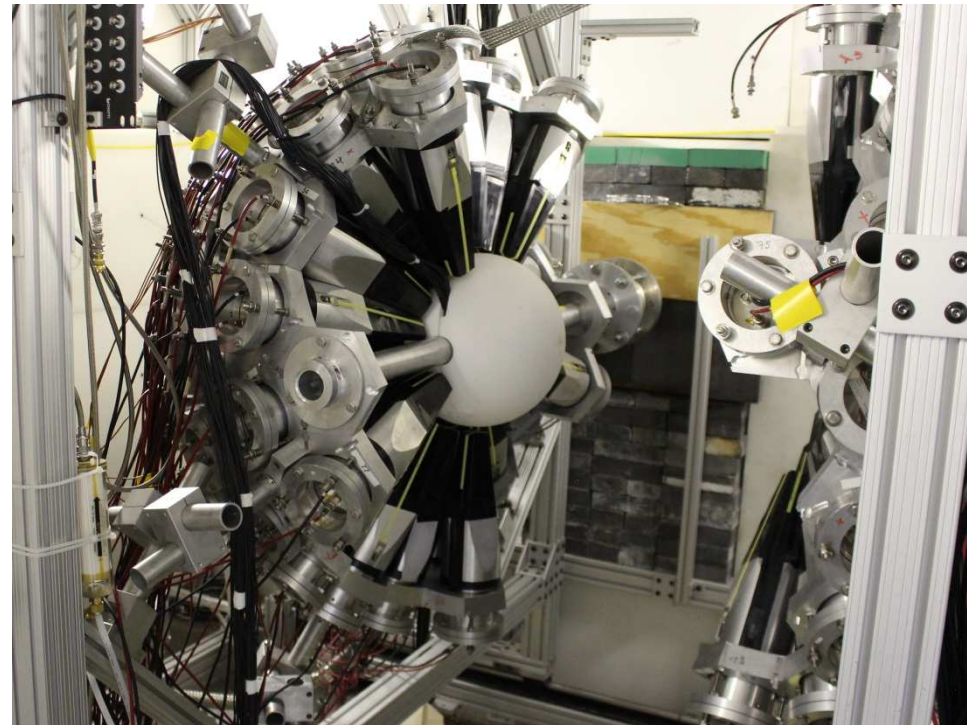
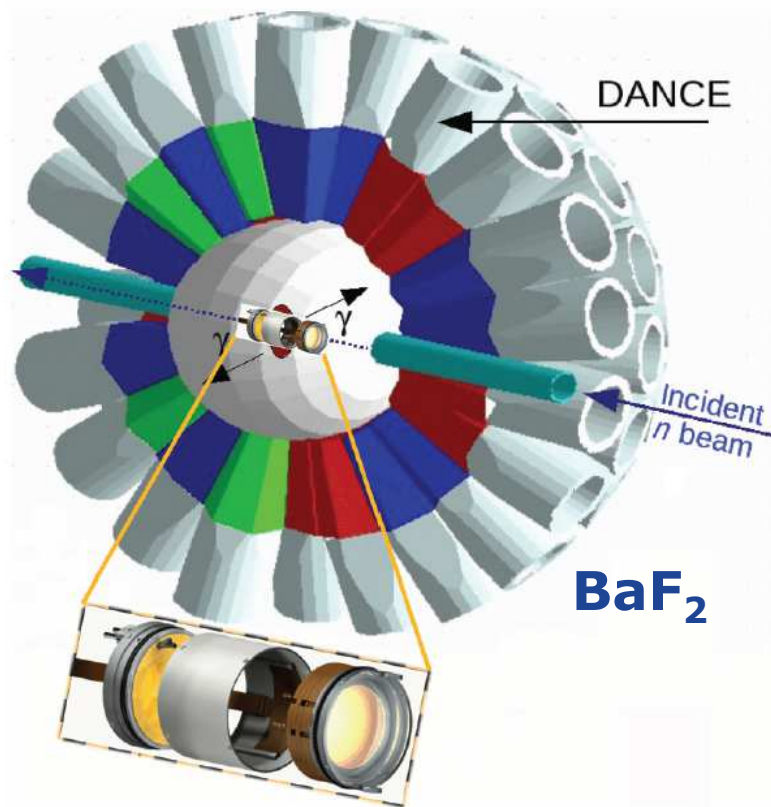
Use of combination of LaBr₃ and HPGe

S. Urlass, Presentation THEORY-4, Varna, 20-22 June 2017

How to measure prompt fission γ -rays

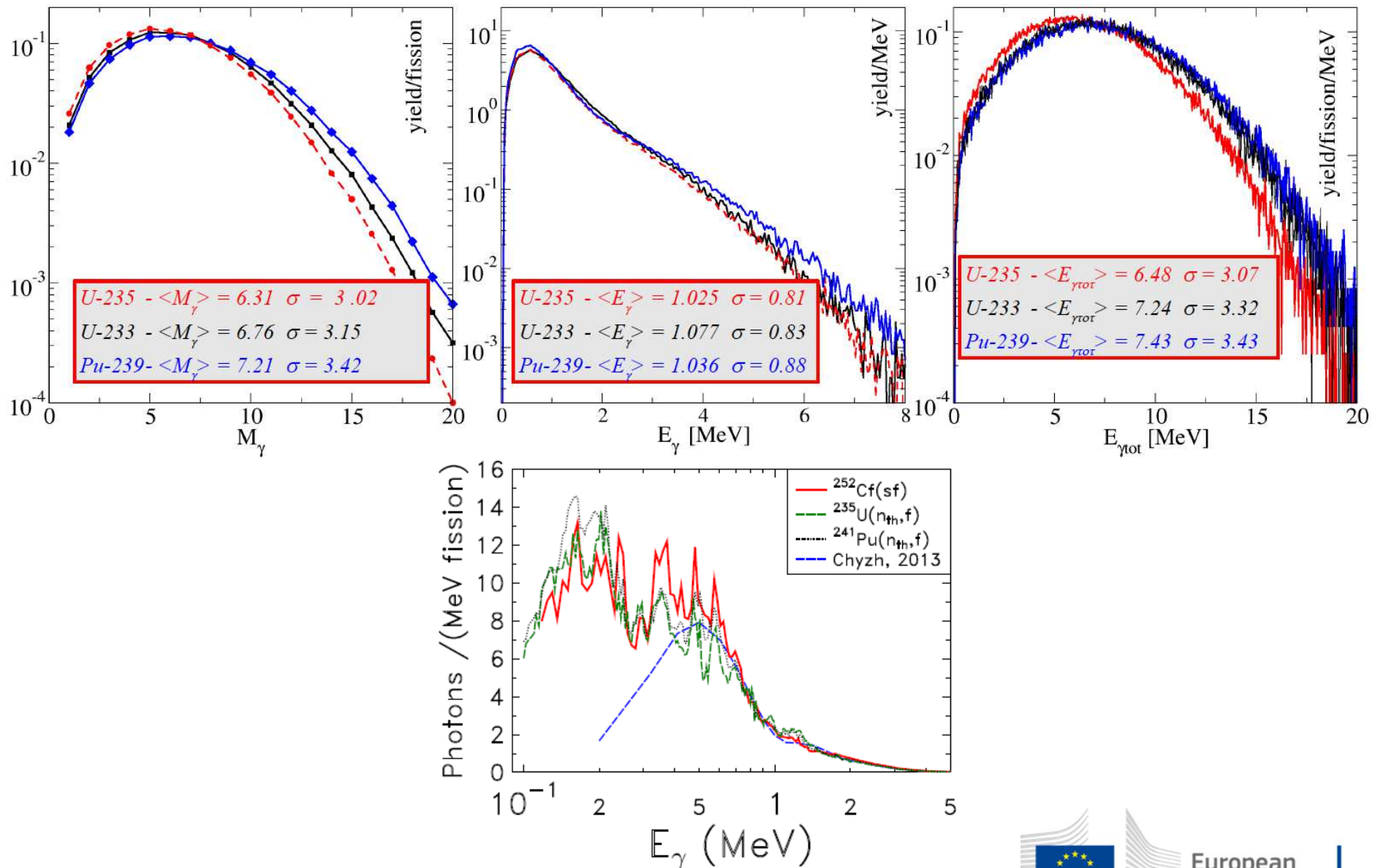
➤ Other devices

➤ DANCE (LANL), 4π array BaF₂ detector array)



How to measure prompt fission γ -rays

M. Jandel, GAMMA-2 Workshop, Sremski Karlovci, RS



S. Oberstedt et al., Rad. Phys. Chem (2017) in press

Literature

In addition to the ones mentioned during the lecture:

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- And many more

**Thank you very much
for Your attention**

